

Resolving Sediment Subduction and Crustal Contamination in the Lesser Antilles Island Arc: a Combined He–O–Sr Isotope Approach

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We report an extensive helium isotope survey of basaltic to andesitic lavas from the Lesser Antilles island arc—an arc system with well-documented evidence of crustal contamination. Given the sensitivity of helium isotopes as a tracer of the effects of crustal additions, our aim is to evaluate the relationship of $^3\text{He}/^4\text{He}$ ratios to other indices of contamination processes such as oxygen and strontium isotopes. To this end, we have carried out 53 $^3\text{He}/^4\text{He}$ analyses on separated minerals (olivines and pyroxenes) from throughout the arc, which we compare with whole-rock strontium and phenocryst oxygen isotope measurements. We show that the three isotopic tracers show coherent patterns throughout the Lesser Antilles, indicating a regional control on crustal contamination. The southern section of the arc (Grenada to Martinique) shows clear evidence for major crustal contamination in all three isotopic systems with results for our samples in the range $^3\text{He}/^4\text{He}_{(\text{olivine})}$ 3.6–7.6R_A, $\delta^{18}\text{O}_{(\text{olivine})}$ 4.74–5.76‰, and $^{87}\text{Sr}/^{86}\text{Sr}_{(\text{whole-rock})}$ 0.703970–0.705463. We suggest that terrigenous sediments incorporated into the arc crust are the principal contaminant. In contrast, there is minimal contamination in the northern part of the arc [Martinique–(Dominica)–Guadeloupe to Saba] with results for the samples in the range $^3\text{He}/^4\text{He}_{(\text{olivine})}$ 6.8–8.4R_A, $\delta^{18}\text{O}_{(\text{olivine})}$ 5.01–5.29‰, and $^{87}\text{Sr}/^{86}\text{Sr}_{(\text{whole-rock})}$ 0.703221–0.703843. In this part of the arc, only pyroxene $^3\text{He}/^4\text{He}$ values (1.4–6.8R_A) consistently record evidence of relatively minor, late-stage additions from the arc crust. Martinique appears

to represent the transition between the two sections of the arc. The tracer with the greatest sensitivity appears to be helium isotopes, as contamination is seen first and most prominently in this system.

KEY WORDS: Lesser Antilles island arc; crustal contamination; helium isotopes; oxygen isotopes; strontium isotopes

INTRODUCTION

Helium isotope variations have found utility as a sensitive tracer of crustal contamination of mantle-derived magmas (Hilton *et al.*, 1992, 1993a, 1993b, 1995; Gasparon *et al.*, 1994; Tedesco *et al.*, 1995; Tedesco, 1996; Patterson *et al.*, 1997). In the Lesser Antilles island arc, Van Soest *et al.* (1998) used $^3\text{He}/^4\text{He}$ ratios of geothermal fluids to argue that crustal contamination plays an important role in petrogenesis in the southern section of the Lesser Antilles but that the central and northern arc shows little or no evidence for crustal contributions. Geothermal fluids, however, give only a broad overview of contamination processes, as intra-island or intra-volcano

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variations in $^3\text{He}/^4\text{He}$ can also develop within the geothermal system as opposed to the magmatic system; this necessitates the adoption of sample selection criteria such as $^3\text{He}/^4\text{He}$ variation as a function of distance from the volcanic edifice (e.g. Sano & Wakita, 1985; Williams *et al.*, 1987; Marty *et al.*, 1989; Hilton *et al.*, 1993b; Van Soest *et al.*, 1998). Helium retentive phenocrysts (e.g. olivine and pyroxene) from lavas offer the potential to circumvent this problem because, in most cases, these minerals can be expected to retain their intrinsic (magmatic) helium since crystallization.

Given the sensitivity of helium isotopes to crustal additions, it is interesting to evaluate the relationship of helium with other indicators of crustal contamination such as mineral composition (e.g. *mg*-number—Gasparon *et al.*, 1994; Hilton *et al.*, 1995; Patterson *et al.*, 1997), oxygen isotopes (e.g. Margaritz *et al.*, 1978; James, 1981; Bacon *et al.*, 1989; Feeley & Sharp, 1995; Macpherson *et al.*, 1998), and strontium isotopes. Therefore, in an attempt to study and better characterize the relationship between helium isotopes and other petrogenetic tracers in subduction zone magmatism, an integrated He–O–Sr isotope study was carried out for the Lesser Antilles using whole-rock powders and olivine and pyroxene phenocrysts. Sr isotopes were determined on the powders, whereas He and O isotopes were determined on olivine and pyroxene separates from the same rocks. We make the case that an integrated approach can provide detailed information about petrogenesis in the Lesser Antilles island arc and the relative role(s) of different styles of crustal contamination. Furthermore, it will be shown that all three isotope systems show a consistent geographical pattern with respect to detecting the presence of crustal contamination, but that, on a sample-by-sample or mineral-by-mineral scale, there is no clear correlation between helium and oxygen or strontium isotopes. For the island of Martinique, a strong correlation between O and Sr isotopes has been shown to exist (e.g. Davidson & Harmon, 1989); this has led to the conclusion that Martinique records more crustal contamination than any other island in the Lesser Antilles. The helium isotope data on mineral separates (presented here) and geothermal fluids (Van Soest *et al.*, 1998; Pedroni *et al.*, 1999) from Martinique suggest that in addition to being a region of major crustal contamination, Martinique also forms the transition between pervasive and probably deep crustal contamination of the southern Antilles and the northern section of the arc where high-level magma chamber contamination predominates.

TECTONIC AND GEOLOGICAL SETTING

Westward subduction of the Atlantic plate has given rise to the Lesser Antilles, an intra-oceanic north–south-trending volcanic arc of 750 km length located on the

eastern margin of the Caribbean plate (Fig. 1). Evidence for current (Montserrat, e.g. Young *et al.*, 1997) to recent (historical to late pre-Columbian times) volcanic activity has been found on most islands from Saba in the north to Grenada in the south (e.g. Maury *et al.*, 1990, and references therein). The oldest dated evidence for volcanic activity in the Lesser Antilles is upper Eocene (Briden *et al.*, 1979). The volcanic arc is split in two from the island of Martinique northwards: an older arc, the Limestone Caribbees [above sea-level expression Grand Terre (Gua-deloupe)–Anguilla], lies to the east of the current Volcanic Caribbees, where active volcanism takes place today (above sea-level expression Martinique–Saba).

The Lesser Antilles arc is characterized by a transition from a typical oceanic arc in the north, with a clearly defined trench of >6 km depth, to a much less typical arc in the south, where a large fore-arc wedge, which reaches a thickness of up to 20 km, completely fills the trench. In the south, the extent of the subduction zone is defined by the deformation front of the fore-arc (e.g. Moore *et al.*, 1982; Westbrook *et al.*, 1988; Fig. 1). The main source of sediments that have contributed to the formation of the large fore-arc wedge and the trench fill in the southern Antilles is the South American continent, via the Orinoco and Amazon river deltas (e.g. Burke, 1988). The thickness of the sediments is greatest in the south, close to the source, and thins towards the north.

At present, the Atlantic plate entering the subduction zone is Campanian or older (Stein *et al.*, 1983; Westbrook *et al.*, 1984). In the south of the arc, the crust may be as old as Jurassic (e.g. Westbrook *et al.*, 1984). Over the history of subduction in the eastern Caribbean, a number of ridges on the Atlantic plate entering the trench have had a significant influence on the subduction process and the shaping of the Lesser Antilles island arc. These ridges, running approximately WNW–ESE, are potentially linked to some of the large transform faults at the Mid-Atlantic Ridge (MAR) (e.g. Roest & Collette, 1986) and are termed ‘transform flanking ridges’ (Stein *et al.*, 1982). They have influenced the Lesser Antilles arc in several ways, not least in the distribution of the sediments in front of the arc: thick in the south as far north as Martinique and then thinning significantly and changing in composition towards the north.

Seismic studies have revealed that the Lesser Antilles arc has a complex crustal structure (Officer *et al.*, 1957, 1959; Bunce *et al.*, 1970). The presence of the large fore-arc and its associated terrigenous sediments in the south, the opening of the Grenada Basin, and the recent arc jump in the north imply that there might be significant differences in the composition of the crust along the strike of the arc (e.g. Fox & Heezen, 1975; Tomblin, 1975; Rea, 1982):

(1) in the south of the arc where the large fore-arc wedge reaches its greatest thickness, it has overthrust the

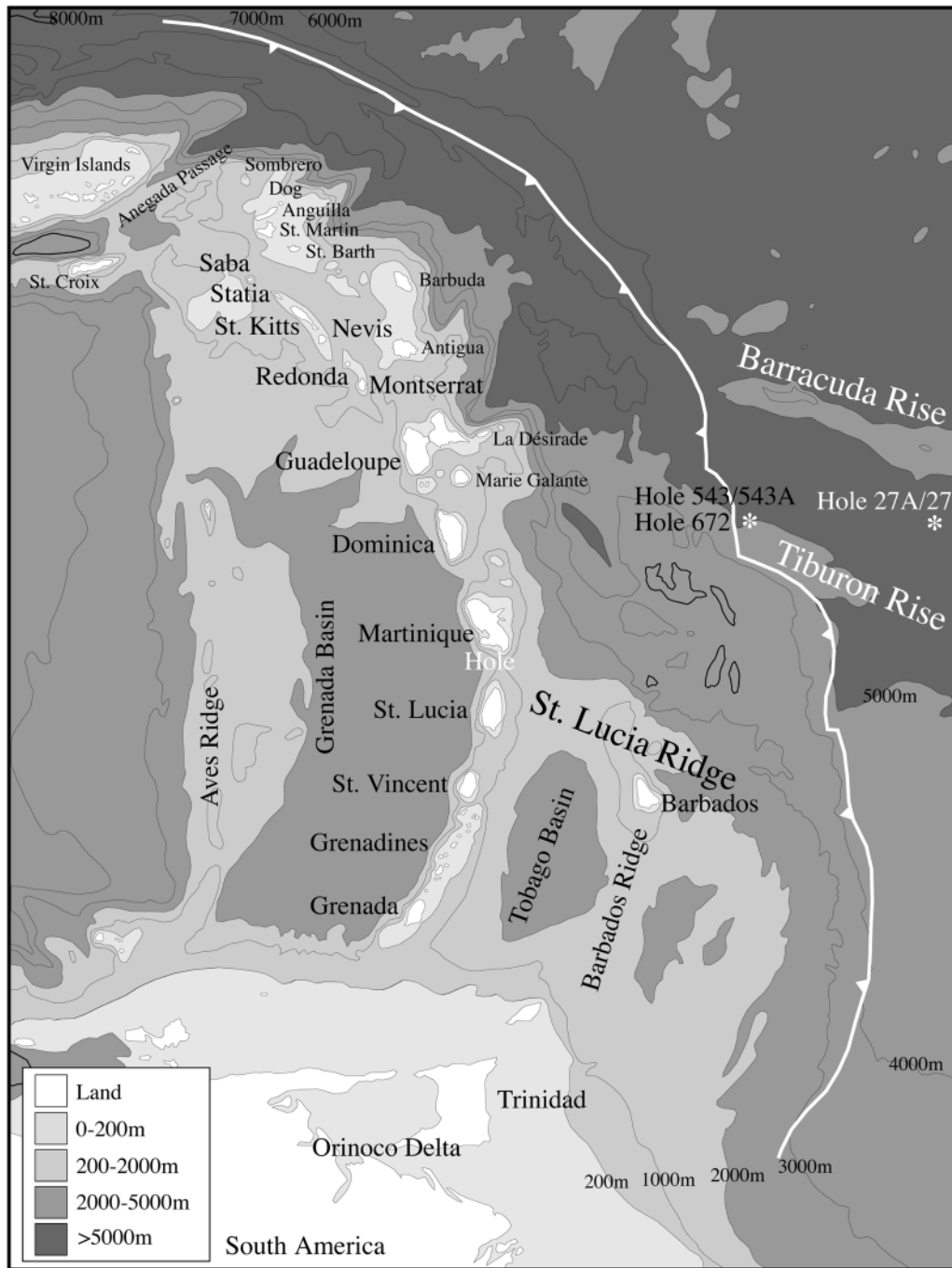


Fig. 1. Map of the Lesser Antilles showing the location of the islands, the trench–deformation front, the main ridges, and the locations of Deep Sea Drilling Project (DSDP) holes 27A/27 and 543/543A and Ocean Drilling Program (ODP) hole 672 (single symbol).

volcanic arc and forms part of the crust (e.g. Torrini & Speed, 1989; Larue *et al.*, 1991). The extent of overthrusting might be related directly to the northward extension of the thick fore-arc. The fore-arc and sediments available for subduction become rapidly thinner north of the St. Lucia Ridge (see bathymetry in Fig. 1,

and, e.g. Ladd *et al.*, 1990). Therefore, it is possible that extension of overthrust fore-arc sediments does not extend beyond St. Lucia or Martinique, where such features have not been described (e.g. Larue *et al.*, 1991).

(2) Whereas the opening of the Grenada Basin in Eocene times produced new oceanic crust in the southern

part of the Lesser Antilles (as far north as Guadeloupe; see Fig. 1), it also caused limited extension of existing arc crust in the north (Bouysse, 1988). Part of the southern arc could therefore be built on relatively young oceanic crust as postulated for some of the Grenadines (Speed & Walker, 1991), whereas the arc north of Martinique could be built on older, pre-Lesser Antilles 'Aves' arc crust, or even on the anomalously thick, oceanic crust of the Caribbean plate. Another possibility is that part of the current southern arc has been built on the remains of the Aves 'fore-arc', which might have been split away from the Aves Ridge when the Grenada Basin opened (e.g. Davidson, 1987; Davidson & Harmon, 1989).

(3) The shift of the volcanic arc north of Martinique (e.g. Westbrook & McCann, 1990) resulted in the northern part of the arc moving away from its original pre-Lower Miocene basement onto the stretched and much thinner older 'Aves' arc or Caribbean plate crust of the northern extension of the Grenada Basin.

These physical changes imply that there are significant changes in crustal composition along the strike of the arc. The effect of the crustal variations on overall crustal thickness along the Lesser Antilles is not completely clear, but a general trend from thicker in the south around St. Vincent (~35–40 km) to thinner in the north around Montserrat (~25 km) can be extrapolated from gravity modeling and seismic data (Maury *et al.*, 1990).

All these features make the Lesser Antilles a complex but well-suited arc for geochemical study. Along-strike changes in crustal composition allow evaluation of the role of crustal contamination and its possible overprinting effects on various subducted components (e.g. Macpherson *et al.*, 1998). On the other hand, the clear differences in type and amount of subducted sediment can be used to better constrain the role and effects of sediment subduction on magmagenesis along the arc (e.g. Arculus & Powell, 1986; Arculus, 1994; Hawkesworth *et al.*, 1994; Pearce & Peate, 1995).

ANALYTICAL TECHNIQUES AND SAMPLES

Analytical techniques

Helium isotope analyses

Samples were processed for $^3\text{He}/^4\text{He}$ and helium abundances ([He]) by crushing *in vacuo* of olivine (ol) and pyroxene (px) [both clinopyroxene (cpx) and orthopyroxene (opx)] separates. The minerals were separated initially by a combination of heavy liquid and magnetic separation using coarsely crushed whole-rock batches. Subsequent hand-picking helped ensure mineral purities close to 100%. Before analysis, the samples were cleaned in analytical grade ethanol.

The majority of the samples were analysed in the Isotope Laboratory of the Scripps Institution of Oceanography (SIO) in La Jolla, California. At SIO, helium analyses were carried out on a 25 cm radius, split-tube, helium isotope mass spectrometer, custom-built for simultaneous collection of ^3He and ^4He (see Rison & Craig, 1983). Gas extractions were carried out using on-line 'solenoid' crushers with released volatiles exposed, in sequence, to a Ti-getter, a charcoal trap held at liquid nitrogen temperature, and a cold-trap cooled by a helium-refrigeration unit to separate the helium from neon. A recent description of the analytical procedure, measurement protocols and typical blanks has been given by Hilton *et al.* (2000).

The remainder of the samples, labeled 'v-' in Table 1, were analysed at the Vrije Universiteit Amsterdam (VUA), which employs a VG T5400 rare gas mass spectrometer, equipped with a Johnston electron multiplier and Faraday cup. The collectors have switchable resistors to facilitate beam measurement over a range of inlet pressures: 10^8 and 10^9 Ω for the multiplier and 10^9 , 10^{10} , and 10^{11} Ω for the Faraday cup. The mass spectrometer is coupled to an all-metal gas preparation line, designed and built in-house, and samples are processed using either on-line pneumatic crushers or an induction furnace. A more complete description of mineral separation procedures, mass spectrometric procedures, and the SIO and VUA crusher types has been given by Van Soest (2000).

Oxygen isotope analyses

Oxygen isotope analyses were performed at Royal Holloway University of London (RHUL) using laser-fluorination (Mattey & Macpherson, 1993). Aliquots of ~1.5 mg of ol, cpx or glass were hand-picked and loaded into the extraction cell (Mattey & Macpherson, 1993). All results were normalized (and corrected if necessary) to two RHUL in-house standards: San Carlos (SC) olivine and GP101 clinopyroxene. These standards are calibrated against a primary standard NIST-30. Accuracy of the analyses is better than 0.1‰ (1 σ). Oxygen yields are >96% for glasses and >98% for mineral separates, thereby ensuring negligible isotope fractionation during fluorination (Mattey & Macpherson, 1993). Data are expressed in the usual δ notation in per mil deviations from the international standard V-SMOW.

Strontium isotope analyses

Strontium isotope analyses were carried out at the VUA on a Finnigan MAT261 multi-collector thermal ionization mass spectrometer. Strontium was prepared using ion exchange columns with Sr-SPEC cation exchange

Table 1: Helium isotope and abundance and oxygen isotope results of phenocryst separates and whole rocks from the Lesser Antilles

Sample no. ^a	Location	Rock type ^b	Wt ^c (g)	mg-no. ^d	R _w /R _A	2σ	[⁴ He] (ncm ³ STP/g)	2σ	δ ¹⁸ O SMOW	⁸⁷ Sr/ ⁸⁶ Sr (WR)	2σ	[Sr] (ppm)
<i>Saba (The Mountain)</i>												
95lss1-ol	Airport	BAS	0.64	88	7.94	0.63	5.60	0.48	—	0.703711	9	278.0
95lss1-gl	Airport	RHY	1.01	21	0.57	0.14	6.71	0.57	—	—	—	—
95lss3-ol	Airport	BAS	1.19	86	6.79	0.52	6.85	0.61	5.29	0.703767	8	249.2
95lss3-gl	Airport	RHY	—	13	—	—	—	—	7.49	—	—	—
96las1-ol	Airport	BAB	1.32	85	7.53	0.37	23.87	1.97	5.13	0.703843	10	320.8
96las1-cpx	Airport	BAB	—	—	—	—	—	—	5.74	—	—	—
<i>Statia (The Quill)</i>												
96lae1-opx	White Wall	AND	1.44	67	6.05	0.45	3.91	0.31	—	0.703601	10	268.6
96lae3-ol	White Wall	MAC	1.17	79	7.53	0.91	0.95	0.09	5.07	0.703567	9	371.9
SE8237-ol	—	BAS	—	—	—	—	—	—	5.34	0.703766 ^e	35	247 ^o
SE8237-px	—	BAS	—	—	—	—	—	—	5.73	—	—	—
SE8240-ol	—	BAS	—	—	—	—	—	—	5.09	0.703723 ^e	23	248 ^o
SE8241-ol	—	BAS	—	—	—	—	—	—	5.00	0.703688 ^e	24	247 ^o
SE8241-px	—	BAS	—	—	—	—	—	—	5.34	—	—	—
SE8245-ol	—	BAS	—	—	—	—	—	—	4.95	0.703632 ^e	22	273 ^o
SE8245-px	—	BAS	—	—	—	—	—	—	5.27	—	—	—
SE8247A-px	—	AND	—	—	—	—	—	—	5.21	0.703759 ^e	27	276 ^o
<i>St. Kitts (Mt. Misery)</i>												
STK104-ol	Fig Tree	BSC	0.91	—	8.03	0.60	7.76	0.63	—	0.703673	12	238.1
95lsk2-ol	Black Rock Cliffs	BAS	1.02	71	8.21	0.61	5.87	0.50	—	0.703661	8	222.3
95lsk2-cpx	Black Rock Cliffs	BAS	1.47	74	5.40	0.71	1.61	0.14	—	—	—	—
95lsk3-ol	Top Mt. Misery	AND	0.92	—	7.70	3.15	0.18	0.04	—	0.703630	8	217.0
95lsk3-cpx	Top Mt. Misery	AND	0.89	70	4.36	0.71	0.33	0.028	—	—	—	—
95lsk3-opx	Top Mt. Misery	AND	0.98	65	6.43	4.23	0.083	0.016	—	—	—	—
<i>Nevis (Nevis Peak)</i>												
95lsn5-opx	Butler's Estate	DAC	2.42	64	1.41	0.86	0.32	0.03	—	0.703548	10	238.3

Table 1: continued

Sample no.: ^a	Location	Rock type ^b	Wt ^c (g)	mg-no. ^d	R _w /R _A	2σ	[⁴ He] (ncm ³ STP/g)	2σ	δ ¹⁸ O SMOW	⁸⁷ Sr/ ⁸⁶ Sr (WR)	2σ	[Sr] (ppm)
<i>Redonda</i>												
v-R8202-ol	—	BAS	0.22	74 ^f	7.51	0.80	1.99	0.12	—	0.703221 ^e	30°	484°
v-R8202-cpx	—	BAS	0.18	79 ^f	6.02	1.70	0.50	0.049	—	—	—	—
v-R8204-ol	—	BAS	0.29	—	8.41	0.74	0.78	0.050	—	0.703243 ^e	34°	493°
v-R8204-cpx	—	BAS	0.24	—	5.64	0.98	0.22	0.037	—	—	—	—
<i>Montserrat (Soufrière Hills)</i>												
95lsmo7-ol	South Soufrière Hill	BAS	0.79	64	8.00	0.93	2.24	0.20	5.12	0.703460	8	488.4
95lsmo7-cpx	South Soufrière Hill	BAS	1.02	73	3.31	0.98	0.70	0.068	—	—	—	—
<i>Guadeloupe (a, Monts Carabes; b, La Soufrière de Guadeloupe)</i>												
GUAD510-ol	—	BSC(a)	0.45	—	7.15	2.17	1.25	0.13	5.005	0.703453	7	260.7
GUAD510-cpx	—	BSC(a)	2.87	—	6.49	0.33	4.85	0.38	5.515	—	—	—
GUAD511-ol	—	BSC(a)	0.48	—	7.49	0.91	4.24	0.38	5.12	0.703500	8	320.8
GUAD511-cpx	—	BSC(a)	2.02	—	6.84	0.29	5.56	0.44	5.35	—	—	—
95lsgu5-cpx	Top Soufrière	AND(b)	1.04	67	3.57	1.92	0.12	0.026	5.12	0.703780	10	195.8
95lsgu5-opx	Top Soufrière	AND(b)	1.48	59	4.41	3.15	0.17	0.020	—	—	—	—
95lsgu5-ol	Top Soufrière	AND(b)	—	81	—	—	—	—	7.12	—	—	—
95lsgu7-cpx	La Regrettee	B-A(b)	1.07	71	4.20	1.41	0.82	0.079	—	0.703337	9	403.9
95lsgu9-opx	Schoelcher	AND(b)	1.18	64	3.79	1.03	0.27	0.034	—	0.703610	7	293.9
<i>Martinique (a, Pitons du Carbet; b, La Montagne Pelée; c, older centers)</i>												
v-M8306-cpx	—	B-A(c)	0.14	—	6.69	0.91	2.31	0.15	—	0.703919 ^f	—	338 ^f
v-95lsm2-cpx	Pitons du Carbet	AND(a)	0.43	—	2.16	0.69	0.53	0.038	6.45	0.705059	10	215.0
v-95lsm4b-opx	Mt. Pelée	AND(b)	0.70	58	3.54	0.41	0.91	0.056	5.69	0.704242	10	268.9
<i>St. Lucia</i>												
v-SL8339-cpx	—	B-A	0.21	—	4.19	1.38	0.55	0.050	—	—	—	—

Sample no.: ^a	Location	Rock type ^b	Wt% (g)	mg-no. ^d	R _{wt} /R _A	2σ	[⁴ He] (ncm ³ STP/g)	2σ	δ ¹⁸ O SMOW	⁸⁷ Sr/ ⁸⁶ Sr (WR)	2σ	[Sr] (ppm)
<i>St. Vincent (Soufrière of St. Vincent)</i>												
STV324-ol	Top Soufrière	BAS	1.04	—	4.72	1.68	0.30	0.04	—	0.703985	9	207.5
STV324-cpx	Top Soufrière	BAS	—	—	—	—	—	—	5.41	—	—	—
v-95ls1-ol	Petit Bordel	BAS	0.43	78	3.56	1.14	1.29	0.08	5.19	0.704447	9	239.9
v-95ls2-ol	Richmond Beach	BAS	0.50	78	6.17	0.54	2.08	0.13	5.36	0.704201	10	197.5
v-95ls4-ol	Top Soufrière	BAS	0.60	70	4.53	1.08	0.51	0.03	4.85	0.703970	10	224.6
v-95ls4-cpx	Top Soufrière	BAS	0.50	73	3.32	0.99	0.47	0.03	5.22	—	—	—
96lav1-ol	Top Soufrière	BAS	1.32	70	4.72	2.54	0.11	0.02	5.36	0.704001	8	225.1
96lav1-ol2	Top Soufrière	BAS	1.32	70	4.72	2.54	0.11	0.02	5.08	—	—	—
96lav1-cpx	Top Soufrière	BAS	0.88	73	4.10	0.50	3.16	0.28	5.47	—	—	—
96lav2-ol	Top Soufrière	B-A	1.17	74	4.40	0.82	1.15	0.11	4.74	0.704114	10	209.5
96lav2-cpx	Top Soufrière	BAS	0.97	73	6.99	1.43	1.35	0.13	4.99	—	—	—
<i>Grenadines (Ile de Caille)</i>												
WIC17-ol	—	BSC	1.11	—	6.28	0.80	2.47	0.21	5.76	0.705194	8	275.4
WIC19-ol	—	BSC	1.15	—	7.02	0.50	21.24	1.81	5.66	0.705463	8	267.5
WIC20-ol	—	BAS	1.08	—	6.54	0.86	4.40	0.39	5.30	0.705208 ^g	23	285 ^g
<i>Kick'em Jenny submarine volcano</i>												
v-KEJ020-cpx	—	B-A	0.23	—	7.90	0.56	5.37	0.32	—	—	—	—
<i>Grenada (a, Northern Domes; b, Mt. Granby–Fedon's Camp; c, young basaltic scoria cones)</i>												
v-GRE27 × 13-ol	—	M-X	0.38	90 ^h	7.64	1.72	0.23	0.028	—	—	—	—
v-GDA138-ol	Radix	BSC(c)	0.50	—	3.60	1.20	0.54	0.04	5.34	—	—	—
v-95ls5b-cpx	Levera Beach	BAS(a)	0.90	77	1.55	0.41	0.77	0.05	5.45	0.704238	12	1245.1
v-95ls5b-ol	Levera Beach	BAS(a)	—	80	—	—	—	—	5.51	—	—	—
v-95ls8-ol	Grand Roy	BAS(b)	0.90	80	4.76	0.79	0.75	0.05	5.64	0.704478	11	998.1
v-95ls8-cpx	Grand Roy	BAS(b)	0.80	77	3.30	0.87	1.87	0.13	5.63	—	—	—
96lag2-ol	Grand Roy	BAS(b)	1.00	81	4.90	0.69	1.65	0.15	—	0.704482	12	1170.0
96lag2-cpx	Grand Roy	BAS(b)	1.19	75	4.82	1.31	1.34	0.14	—	—	—	—
96lag3-cpx	Point David	AND(a)	0.76	75	4.99	1.45	0.39	0.06	—	0.705154	6	456.7
96lag4-ol	D'Arbeau Hill Quarry	BSC(c)	1.18	87	7.51	0.55	14.97	1.21	5.18	0.705082	6	296.0

^aPrefix 'v-' indicates He isotopes of sample measured at the Vrije Universiteit Amsterdam; all others measured at Scripps Institution of Oceanography; 95 or 96 indicates year of field season in which sample was collected. ol, olivine; cpx, clinopyroxene; opx, orthopyroxene; gl, glass; wr, whole rock. Sample abbreviation in capitals indicates sample obtained from third parties (see Acknowledgements).

^bBAS, basalt; B-A, basaltic andesite; AND, andesite; DAC, dacite; RHY, rhyolite; BAB, basaltic bomb; BSC, basaltic scoria; MAC, mafic cumulate; M-X, mantle xenolith.

^cWeight of crushed sample in grams.

^dmg-number = $\text{Mg}^{2+}/(\text{Mg}^{2+} + \text{Fe}^{2+}) \times 100$.

^eData from Davidson (1987)

^fData provided by Jon Davidson of the University of Durham, UK.

^gData from White & Dupre (1986)

^hData provided by Cheryl Williams of the Open University, Milton Keynes, UK.

resin (100–150 µm). A procedural blank was determined with each batch of samples; typically, this blank ranged from 100 to 300 pg Sr. Samples were loaded on annealed Re single filaments, with Sr isotope ratio measurements made using the Sr triple-jump method: this procedure corrects on-line for mass fractionation of the ratio. Each turret of (13) samples contained at least one aliquot of the NBS987 Sr standard. A complete description of clean laboratory and mass spectrometric procedures has been given by Van Soest (2000). Strontium contents of the samples were determined by inductively coupled plasma–mass spectrometry (ICP-MS).

Samples

The majority of samples used for this study were phyrlic lavas of basaltic to andesitic composition. During field collection, emphasis was placed on collecting olivine-bearing samples given the propensity of pyroxenes to contain He with a lower $^3\text{He}/^4\text{He}$ ratio than cogenetic olivine [see below and Hilton *et al.* (1995)]. Also, we attempted to sample ‘young’ lava flows based mainly upon field relationships and published age data (e.g. Briden *et al.*, 1979; Maury *et al.*, 1990). All samples were analysed for major and trace elements by X-ray fluorescence spectroscopy and ICP-MS, respectively, and mineral compositions were determined by microprobe. These results have been discussed by Van Soest (2000), and will be reported elsewhere. Rock type and *mg*-number are given in Table 1.

RESULTS

Helium isotope results

A total of 53 helium isotope and abundance analyses were carried out on mineral separates from 38 samples covering 12 islands and one submarine volcano. Twenty-six ol and 26 px separates (21 cpx and five opx) were analyzed. Additionally, one rhyolitic glass (gl) separate was processed. Results are reported in Table 1. Helium isotope ratios ($^3\text{He}/^4\text{He}_{\text{sample}} = R_M$) are reported in the R_M/R_A notation, where R_A is the isotopic ratio of helium in air. Helium abundances are reported in $\text{ncm}^3 \text{STP/g}$. Neon was measured for some samples; however, this was not possible in all cases as a result of low gas contents. Therefore, a correction was not made for potential air contamination of the samples, as is usually performed for helium analyses of geothermal fluids. It is significant that in the case of samples where an air correction was possible, shifts in $^3\text{He}/^4\text{He}$ ratio were typically $<0.1R_A$; this gives confidence that the effects of air contamination of phenocrysts are negligible.

Helium isotope results obtained by crushing of ol and px show all samples to be characterized by a significant mantle-derived component. The highest values overlap with mid-ocean ridge basalt (MORB) helium ($8 \pm 1R_A$), as observed for a number of other arcs worldwide (e.g. Poreda & Craig, 1989; Gasparon *et al.*, 1994), whereas minimum ratios of $1.4R_A$ (Nevis Peak) imply a large radiogenic helium contribution. The ratio of $0.57R_A$ in the crushed rhyolitic glass implies an even larger radiogenic helium contribution. Although we consider most samples to be young ($<1 \text{ Ma}$), disturbance of the original magmatic $^3\text{He}/^4\text{He}$ ratio is a concern. This could occur by addition of radiogenic ^4He , as a result of decay of U- and Th-series radionuclides, or by in-growth of cosmogenic ^3He by exposure at or near the surface. In either case, the extraneous helium is probably sited in the mineral matrix and not in fluid and/or melt inclusions trapped in the mineral lattice. Processing samples by vacuum crushing, particularly if crushing times are minimized (as in this work; see also Hilton *et al.*, 1993b; Scarsi, 2000), largely overcomes the problem of addition of extraneous helium as only inclusion-sited volatiles are released. We conclude, therefore, that the possible contribution of a radiogenic or cosmogenic helium component to the magmatic helium released by crushing is minimal.

In Fig. 2a, the helium isotope results are plotted against position (latitude) along the arc. If only the ol data (black symbols) are considered, we can observe two distinct groups: a northern group of islands [Guadeloupe, Montserrat, Redonda, (Nevis), St. Kitts, Statia, and Saba], characterized by MORB-like $^3\text{He}/^4\text{He}$ ratios, and a southern group of islands (Grenada, Grenadines, and St. Vincent) with a larger range in $^3\text{He}/^4\text{He}$ values ($3.56\text{--}7.94R_A$), which is generally lower than MORB. The three central islands (St. Lucia, Martinique, and Dominica) do not have suitable ol-bearing samples and therefore cannot be classified in this way.

Pyroxene data have a large range in $^3\text{He}/^4\text{He}$ ratios throughout the arc (white and gray symbols in Fig. 2a). It is significant that for the northern group of islands most px data are lower than the ol data, whereas for the southern group px and ol data overlap, with px extending to lower values. The px data for the adjacent islands St. Lucia and Martinique again do not allow classification of these islands into the two geographic groupings.

If the ol helium isotope results are compared with results obtained from geothermal fluids [diagonally hatched boxes in Fig. 2a; data from Van Soest *et al.* (1998)], it is clear that considerable overlap exists between the two datasets. The mineral data of the central group of islands, comprising Martinique, Dominica, Guadeloupe and Montserrat, place these islands within the domain of the northern portion of the arc, i.e. $^3\text{He}/^4\text{He}$ within error of the MORB value. In contrast, St. Lucia appears

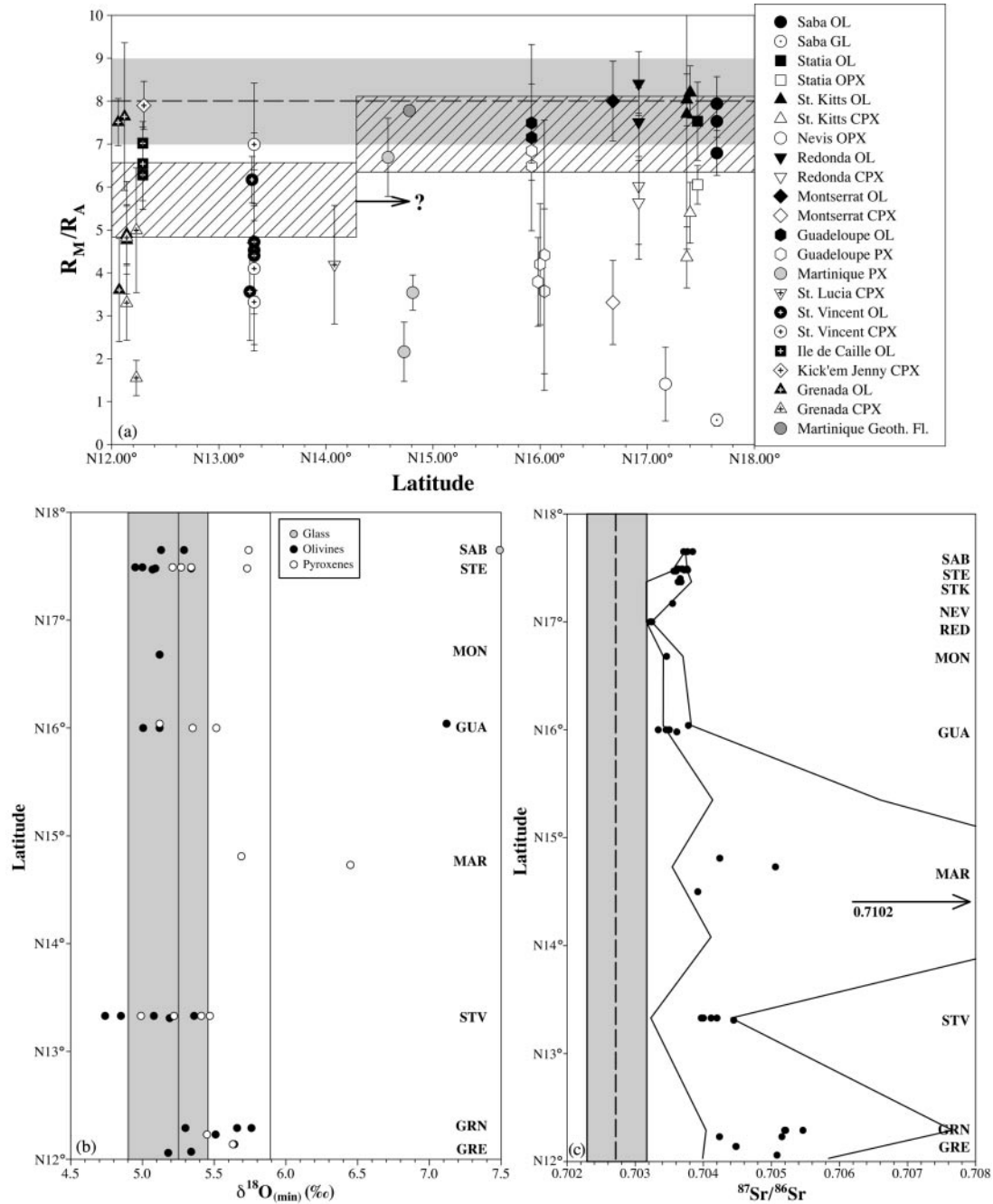


Fig. 2. An overview of the helium, oxygen, and strontium isotope results vs location (latitude) in the arc. (a) R_M/R_A vs latitude for the complete mineral database for the Lesser Antilles. The gray box shows the MORB range and the two cross-hatched boxes show the range for Lesser Antilles geothermal fluids in both parts of the arc (Van Soest *et al.*, 1998). Data are shown for each island and for mineral phase analysed. (b) $\delta^{18}O$ vs latitude for the complete mineral dataset obtained during this study. Data are shown per island and mineral type. Also shown are two boxes for the range of olivine (gray box) and clinopyroxene in equilibrium with mantle peridotite (Mattey *et al.*, 1994). GRE, Grenada; GRN, Grenadines; STV, St. Vincent; MAR, Martinique; GUA, Guadeloupe; MON, Montserrat; STE, St. Eustatius; SAB, Saba. (c) $^{87}Sr/^{86}Sr$ in WRs plotted against position in the arc (latitude). The MORB range is indicated by the gray box and is based on values from Saunders *et al.* (1988) and Rollinson (1993, and references therein). The two continuous lines indicate the range of published Sr isotope ratios for the Lesser Antilles [see text and Van Soest (2000) for references]. RED, Redonda; NEV, Nevis; STK, St. Kitts. The arrow marked by 0.7102 indicates the highest $^{87}Sr/^{86}Sr$ determined for the island of Martinique.

to belong to the southern group. For Martinique, the situation in helium isotope systematics is ambiguous, as $^3\text{He}/^4\text{He}$ span a wide range; indeed, the helium results for Martinique [Table 1 and Van Soest *et al.* (1998) and Pedroni *et al.* (1999)] suggest that it could form the transition region between the two groups. This issue will be addressed further below.

Oxygen isotope results

Oxygen isotope data for 41 mineral separates (24 ol, 16 px, and one gl sample) are reported in Table 1. Helium isotope and abundance data are available on all but 13 of these samples. In Fig. 2b, the oxygen data are plotted against geographical position in the arc. For reference, we also show the ranges of oxygen values for ol and cpx in equilibrium with mantle peridotite (Mattey *et al.*, 1994). Most data for the Lesser Antilles fall within the respective ranges for mantle peridotite. This suggests that these minerals crystallized from a melt that had undergone little or no modification of its oxygen isotope systematics since its formation in the mantle wedge.

However, there are 12 samples that fall outside the range of mantle peridotite. It is noteworthy that, of these, nine come from the south of the arc (Grenada–Martinique). There are two outliers, showing large ($>0.5\%$) deviations from mantle peridotite values: Martinique (lsm2-cpx) and Guadeloupe (lsgu5-ol). Petrographic and microprobe studies suggest that the lsgu5-ol separate is xenocrystic and therefore might not be representative of the magma. On the other hand, the lsm2-cpx separate appears to be phenocrystic and thus representative of the magmatic $\delta^{18}\text{O}_{\text{cpx}}$ value.

The other 10 samples (four cpx and six ol) show deviations from mantle peridotite at the level of $\sim 0.3\%$ or less. This suggests that these mineral separates crystallized from a magma that had modified oxygen isotope systematics compared with mantle peridotite. Within this sub-set, six samples (four cpx and two ol) deviate towards lower $\delta^{18}\text{O}$ values (as low as 4.74% for ol and 4.99% for cpx). Four of these six samples come from the island of St. Vincent, and form two ol–cpx mineral pairs (lsv4 and lav2); the other samples are cpx separates from Guadeloupe (lsgu5-cpx) and Statia (SE8247A). The remaining four samples—all ol separates—show small deviations towards higher $\delta^{18}\text{O}$ values (up to 5.76%): two from Ile de Caille (WIC17 and WIC19) and two from Grenada (lsg5 and lsg8). The glass separate from Saba (lss3-gl) has the highest $\delta^{18}\text{O}$ value obtained in this study. The $\delta^{18}\text{O}$ value of 7.49% (vs SMOW) is consistent with a glass of rhyolitic composition that has evolved in the arc crust (e.g. Harmon *et al.*, 1984; Ellam & Harmon, 1990).

The oxygen isotope database contains 12 ol–cpx mineral pairs. Except for three samples (lsgu5, lsg5, and lsg8)

the mineral pairs have $\Delta_{\text{ol-cpx}}$ values that fall within the range of mantle peridotite (Mattey *et al.*, 1994). The lsgu5 pair has a much larger $\Delta_{\text{ol-cpx}}$ value, probably because the ol in the sample is xenocrystic although the cpx is also offset but towards lower values. The other two pairs (lsg5, lsg8) have $\Delta_{\text{ol-cpx}}$ values that are less than the mantle peridotite range. This is caused by significantly higher ol $\delta^{18}\text{O}$ values compared with mantle peridotite, whereas cpx values have remained within the peridotite range (Mattey *et al.*, 1994).

Two of the mineral pairs of St. Vincent (lsv4 and lav2) have both ol and cpx $\delta^{18}\text{O}$ values that are offset compared with mantle peridotite, but $\Delta_{\text{ol-cpx}}$ values that remain within the mantle range. This indicates that both ol and cpx have been offset by the same amount. Another interesting feature of St. Vincent is the lav1 ol sample, which has two distinct $\delta^{18}\text{O}$ values. On the basis of the $\Delta_{\text{ol-cpx}}$ systematics of the other mineral pairs, it seems that the olivine with the higher value of 5.36% could be of xenocrystic or cumulate origin.

In comparison with the helium isotope data (Fig. 2a), the oxygen isotope data in Fig. 2b do not show any clear systematic inter- or intra-island trends. It is interesting to note, however, that of the 12 samples that do show deviations from mantle peridotite values, eight are from the south of the arc (Grenada–St. Vincent) and one is from Martinique. This is exactly the section of the arc where helium isotopes in olivines (Fig. 2a) and geothermal fluids (Van Soest *et al.*, 1998; Pedroni *et al.*, 1999) deviate from MORB values. This is also the section of the arc for which the presence of crustal contamination has been advocated previously (Thirlwall & Graham, 1984; Davidson, 1987; Davidson & Harmon, 1989; Smith *et al.*, 1996; Thirlwall *et al.*, 1996).

Strontium isotope results

A total of 30 whole-rock (WR) samples were analysed for $^{87}\text{Sr}/^{86}\text{Sr}$ at the VUA. WR Sr isotope ratios cover a range from 0.703221 (R8202) to 0.705463 (WIC19) (Table 1). This wide range is mainly found in the south of the arc—the islands of Grenada, the Grenadines, and Martinique (0.703919–0.705463). The northern arc, from Guadeloupe northwards, is characterized by a much narrower range (0.703221–0.703843). Minimum values for $^{87}\text{Sr}/^{86}\text{Sr}$ are found at the islands of Montserrat and Redonda. Sr contents generally range between ~ 200 and ~ 350 ppm, but at three islands, much higher abundances are observed: Grenada has rocks with Sr concentrations up to ~ 1250 ppm, and the islands of Montserrat and Redonda have values around 490 ppm.

In Fig. 2c, the data are plotted against position (latitude) in the arc. Also shown is the range of previously reported $^{87}\text{Sr}/^{86}\text{Sr}$ for the Lesser Antilles. It can be seen that the

range of literature $^{87}\text{Sr}/^{86}\text{Sr}$ for the southern arc is much larger than that found in this study, whereas there is good overlap for the northern arc. This effect is probably due to sampling bias and the geographical distribution of rock types. In this study, the focus was mainly on ol-bearing rocks so that more evolved rocks, which are mainly responsible for the high $^{87}\text{Sr}/^{86}\text{Sr}$ in the south, were not sampled. Additionally, these more evolved rock types are much less abundant in the northern part of the arc, which helps explain the good overlap between our data and literature values.

It is noteworthy that most $^{87}\text{Sr}/^{86}\text{Sr}$ data (from this study and the literature survey) are significantly higher than the range reported for MORB (e.g. Saunders *et al.*, 1988). In Fig. 2c, the MORB range is shown as a gray box and the data fall within this range only for the island of Redonda. In general, the northern arc (Guadeloupe–Saba) has $^{87}\text{Sr}/^{86}\text{Sr}$ similar to other oceanic arcs worldwide (e.g. White & Patchett, 1984; Woodhead, 1989; Elliott *et al.*, 1997; Turner *et al.*, 1997), whereas the southern arc (Grenada–Martinique) trends towards much higher radiogenic ‘crustal’ values (especially noticeable in the literature range) similar to continental arcs such as the Andes (e.g. McMillan *et al.*, 1993). A notable exception to this generalization is the narrow range in Sr isotope ratios for the island of St. Vincent in the southern arc, which appears very similar to the Sr isotope systematics of the northern arc.

Finally, we note that the occurrence of high or radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ in the southern Lesser Antilles coincides with the region of low $^3\text{He}/^4\text{He}$ ratios in ol and geothermal fluids, and with the majority of the $\delta^{18}\text{O}$ values that deviate from the MORB range.

DISCUSSION

Helium isotope systematics and helium contents

The helium isotope systematics of the phenocrysts (this study) conform to the general He pattern derived from our earlier survey of geothermal fluids, i.e. MORB-like $^3\text{He}/^4\text{He}$ ratios in the northern Antilles with a pronounced radiogenic He addition in the south (Van Soest *et al.*, 1998). The phenocryst database is more extensive, however, and allows for closer scrutiny of the details of the distribution, particularly the location of the transition where radiogenic helium starts to contribute to the volatile inventory.

In Fig. 3, we plot $^3\text{He}/^4\text{He}$ vs helium content ([He]) for the entire database of ol and cpx separates of the present study. For the northern islands (Fig. 3a), the olivine $^3\text{He}/^4\text{He}$ data do not show any correlation with [He], whereas the pyroxene isotope data show a clear decrease with decreasing [He] (limiting case shown by

the black arrow). The Martinique data seem to follow this latter trend. Significantly, the southern arc does not show the same trends. In this case (Fig. 3b), the data are more scattered, with significantly lower than MORB helium isotope ratios characterizing the majority of the ol and px samples.

We consider several factors that may cause these various features: (1) magma aging in crustal magma chambers; (2) modification of the mantle wedge, either by subduction of helium directly or by enhanced in-growth as a consequence of increased U and Th contents associated with subduction; (3) isotopic fractionation during batch or fractional degassing; (4) degassing, combined with either a localized late-stage assimilation or a major crustal contamination event.

(1) The possibility of in-growth of ^4He in crustal magma chambers (or magma aging) as the cause of $^3\text{He}/^4\text{He}$ ratios lower than MORB in arc-related lavas was considered by Hilton *et al.* (1993b) for the Andes, with the conclusion that the expected residence times of magmas in the crust are too short to produce sufficient radiogenic helium to modify magmatic $^3\text{He}/^4\text{He}$ values. Using the same approach, we calculate that for an initial MORB-like helium composition ($^3\text{He}/^4\text{He} = 8R_A$, [He] = $15 \times 10^{-6} \text{ cm}^3 \text{ STP/g}$; Sarda & Graham, 1990) it would take a residence time of ~ 20 my to grow in the requisite $2.14 \times 10^{-6} \text{ cm}^3 \text{ STP } ^4\text{He}$ to lower the $^3\text{He}/^4\text{He}$ ratio by one R_A unit (for an average Th and U composition of basaltic and basaltic–andesitic lavas in the Lesser Antilles of [U] = 0.6 ppm, Th/U = 2.5; Van Soest, 2000). Magma residence times of up to 60 ky have been calculated for evolved magmas from St. Vincent based on inferred isochronous relationships in U-series isotopes from minerals in recent lavas (Heath *et al.*, 1998b). Even shorter residence times are anticipated based on the presence of Ra disequilibria in recent lavas from Martinique, St. Vincent and Kick’em Jenny (Chabaux *et al.*, 1999). Such short crustal residence times inferred for Lesser Antilles magmas are grossly insufficient for magma aging to affect magmatic $^3\text{He}/^4\text{He}$ values.

(2) The presence of radiogenic helium in the southern Lesser Antilles has been explained by the subduction of radiogenic helium acting to modify the mantle wedge (Pedroni *et al.*, 1999). This conclusion was based upon a tentative correlation between helium isotope systematics in geothermal fluids and strontium isotope systematics from published whole-rock samples. As discussed below, we consider this approach untenable. However, we also point out that the high diffusivity of helium in deep-sea sediments (Hiyagon, 1994a, 1994b) makes it highly improbable that helium will be retained until sediment reaches the zone of magma generation; indeed, it is far more likely that helium will be lost to pore fluids during burial and the initial stages of subduction (Hilton *et al.*, 1992).

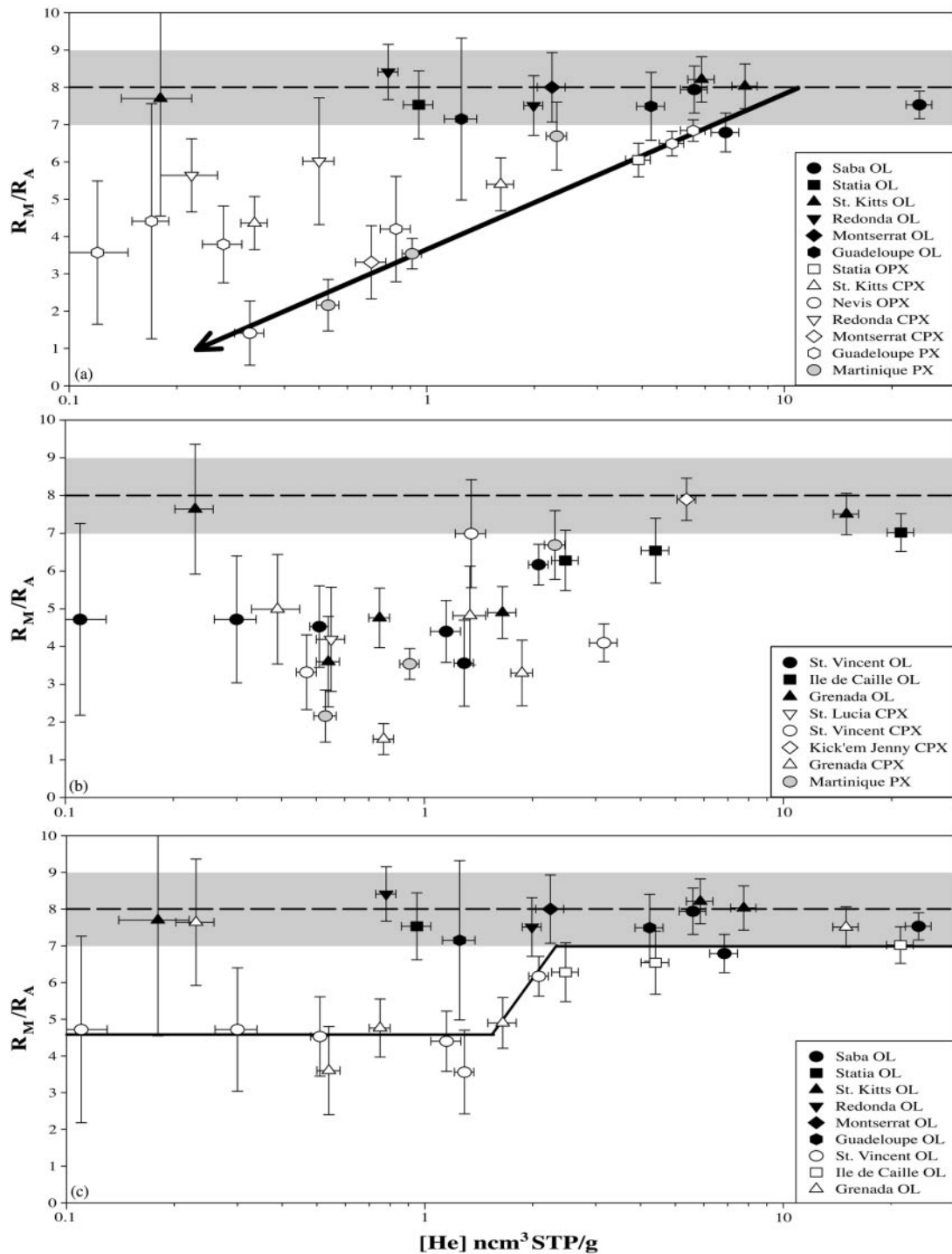


Fig. 3. (a) A plot of R_M/R_A vs $[He]$ for the northern arc. Data are shown for island and mineral type. There are two clear trends: ol $^3\text{He}/^4\text{He}$ data show no correlation with helium contents and are characterized by MORB-like $^3\text{He}/^4\text{He}$ for the complete range of helium contents. In contrast, the northern arc px $^3\text{He}/^4\text{He}$ data show a clear positive correlation with helium content. The data for Martinique seem to conform to the trend observed for the northern arc px data. (b) A plot of R_M/R_A vs $[He]$ for the southern arc. Data are shown per island and per mineral type. Trends are less obvious than in the case of the northern arc px data, although there appears to be a broad positive correlation of most samples (including the olivines) with helium content. (c) A plot of R_M/R_A vs $[He]$ for ol data from the whole arc. Data are shown per island. This plot illustrates: (1) the marked difference in ol data obtained from the two segments of the arc; (2) the fact that the southern ol data do not define a correlation between helium isotope ratio and helium content—rather there is a horizontal trend with a step-function to the northern data. This horizontal trend is similar to the trend of the northern ol data but displaced to lower ratios.

In-growth of radiogenic helium in the mantle wedge could also result from increased amounts of U and Th introduced into the mantle by subduction. Again, however, this mechanism cannot explain our observations for the Lesser Antilles, as we observe $^3\text{He}/^4\text{He}$ ratios equal to the MORB-mantle value ($8R_A$) throughout the arc, including a mantle xenolith from Grenada in the south of the arc (GRE27x13-ol). Furthermore, short residence times of U and Th series isotopes in the mantle wedge argue against this possibility; for example, based on Ra disequilibria observed in lavas from the Lesser Antilles, Chabaux *et al.* (1999) concluded that melt formation and extraction must be rapid—within 8 ky.

The fact that it is highly improbable that helium will be subducted in significant amounts suggests that any significant in-growth in the wedge, even at elevated U and Th contents, would be of similar if not longer time scales compared with the results of the calculations under point (1). This suggests that magmas throughout the Lesser Antilles will enter the crust with MORB-like helium isotope ratios.

(3) Isotopic fractionation of He during degassing may potentially cause lowering of $^3\text{He}/^4\text{He}$ in residual volatiles. Theoretically, if diffusion controlled, the effect is small, $\sim 15\%$ (Hilton, 1996). However, it is unlikely that such an effect occurs in nature. Kurz & Jenkins (1981), for example, noted that $^3\text{He}/^4\text{He}$ of vesicle and host glass were equal even though helium was overwhelmingly partitioned into the vesicle phase.

(4) Degassing can play a pivotal role in determining the helium isotope systematics of the phenocrysts, not by isotopic fractionation, but by reducing intrinsic helium contents of magma thereby increasing its susceptibility to crustal contamination (Hilton *et al.*, 1993b). Using the He partition coefficient of 0.008 between olivine and MORB melt (Marty & Lussiez, 1993), it would be expected that an olivine phenocryst that crystallized from a MORB magma ($15 \times 10^{-6} \text{ cm}^3 \text{ STP/g}$) would contain $1.2 \times 10^{-7} \text{ cm}^3 \text{ STP/g}$ helium. Our observed range in helium abundances (Table 1) is considerably lower than this value (approximately one order of magnitude for the highest concentration observed: $\sim 2.5 \times 10^{-8} \text{ cm}^3 \text{ STP/g}$). This implies that if the observed He concentrations do indeed represent equilibrium partitioning between olivine and magma then equilibrium must have occurred with a magma that is degassed relative to pristine MORB. The inferred high state of degassing implies that addition of relatively small amounts of radiogenic helium from the crust could greatly affect the helium isotope ratios.

This last mechanism appears the most plausible explanation for the observed $^3\text{He}/^4\text{He}$ characteristics of the Lesser Antilles. The observation of generally lower $^3\text{He}/^4\text{He}$ in px throughout the arc is particularly relevant, and can be explained by the higher He diffusion coefficient of px compared with ol (Trull & Kurz, 1993). In this way,

crustal helium will be preferentially incorporated into px and result in lower $^3\text{He}/^4\text{He}$ than cogenetic ol. We test this conclusion below by considering the relationship between $^3\text{He}/^4\text{He}$ and mineral chemistry.

Geographical control on crustal contamination

The previous section has identified two distinct trends in the helium isotope data. In the northern section of the arc, px has preferentially recorded the addition of radiogenic helium. No such evidence is found in ol even if helium contents are low. In contrast, both ol and px record evidence for crustal additions in the southern section of the arc. In this section, we suggest that this observation records difference in the circumstances of contamination—the timing of crustal additions and possibly the nature or availability of the contaminant.

In the northern arc, ol does not record the crustal addition, indicating that ol has already ceased to be a major crystallizing phase before any contamination: in effect, the ol has become closed to helium exchange with the host magma. However, because px has lower $^3\text{He}/^4\text{He}$, we infer that the crustal contamination must have occurred at a late stage in the evolution of the magma. This is because (1) contamination must occur following degassing (which is exacerbated at low pressures, i.e. shallow levels in the crust), and (2) px has higher helium diffusivities than ol (Trull & Kurz, 1993) and therefore will remain ‘open’ for exchange with the melt at lower temperatures and/or for longer time periods. The most plausible process to add radiogenic helium preferentially to px would be late-stage assimilation of, or interaction with, older arc crust. Such crust would be expected to have grown-in radiogenic helium and thereby possess the requisite contaminant signal.

In the southern arc, most ol phenocrysts record addition of radiogenic helium implying that ol was still crystallizing or still open to exchange with the host magma during the contamination process. In detail, however, the ol data (Fig. 3c) appear to define a more or less horizontal trend ($\sim 4.7R_A$) in the southern arc with a step function towards higher, MORB-like helium isotope ratios at higher helium contents. This step in the data is also reflected in the WR major element chemistry and in the phenocrysts: the high $^3\text{He}/^4\text{He}$ and [He] lavas have markedly higher WR MgO contents and ol *mg*-number than the other southern arc samples (Van Soest, 2000). We refer to this group of samples as the ‘high *mg*-number’ group samples, and they show little evidence for crustal contamination in their helium isotope systematics. The remaining samples form a horizontal trajectory in Fig. 3c, similar to that for the ol data from the northern arc but clearly displaced to lower $^3\text{He}/^4\text{He}$. We interpret this

observation as a regional-scale contamination process that must have taken place before or during ol crystallization. We note that this process has also affected the geothermal fluids; this implies that the contaminant must be a major source of radiogenic helium and that contamination must have taken place early in the degassing history of the magmas. Contamination from this source modified the helium isotope ratios of the southern arc magmas from MORB-like values to $\sim 4.7R_A$ for the olivines and, on average, to slightly higher values for the geothermal fluids.

Following the addition of radiogenic helium, we suggest that the ol crystallized normally and became closed for helium exchange. Subsequently, the magmas could have been affected by late-stage crustal assimilation although its presence is less obvious than in the north. This is probably a consequence of the fact that radiogenic helium would have been added early in the magmatic history leaving less leverage for isotopic modification by late-stage assimilation.

The contrast between the contamination history of the northern Antilles and that of the southern portions of the arc is illustrated further in Fig. 4, in which helium isotope ratios of mineral pairs from the same sample are compared. If magmas were unaffected by contamination processes then all mineral pairs should be in equilibrium and lie in the MORB field (shaded square)—this is clearly not the case. The northern suite shows disequilibrium, with px having consistently lower $^3\text{He}/^4\text{He}$ than cogenetic ol; in this case, we conclude that the helium isotope ratio was modified late in the evolution of the magma. The southern suite has ol and px $^3\text{He}/^4\text{He}$ close to equilibrium but clearly displaced from the MORB field: we interpret this as evidence for early-stage, pervasive crustal contamination. The slight He-isotope disequilibrium for some of these samples, with px lower than ol, shows that these magmas also have been affected by late-stage assimilation, but that the magmatic $^3\text{He}/^4\text{He}$ is dominated by the early contamination events.

One of the prime candidates for the crustal contaminant in the southern Antilles is the presence of terrigenous sediments in the arc crust. These lithologies may be overthrust sediments of the present fore-arc or 'fore-arc' sediments from the older Aves arc that was split during the opening of the Grenada back-arc basin. Both are likely to contain large amounts of old terrigenous sediments from the South American continent. These deposits are not expected to extend much further north than Martinique. Another potential reservoir for the radiogenic helium could be the arc crust itself, which is significantly older in the south of the arc. However, it is unclear if sufficient helium could have been produced by radioactive decay in the crust since the Eocene—assuming that this crust degassed during formation. The relevant factors are the radio-element content of the

arc crust, its retention characteristics, and the helium concentration contrast between the crust and the intruding magma. Arguments based on the Sr and Pb isotope composition of typical arc crust vs more evolved Lesser Antilles lavas (Thirlwall & Graham, 1984; Davidson, 1987; Davidson & Harmon, 1989; Smith *et al.*, 1996; Thirlwall *et al.*, 1996) indicate that the arc crust could not be the sole reservoir responsible for the crustal contamination events.

The location of Martinique with respect to the two geographical groupings remains unclear. Taking into account the fact that the Martinique px data follow the trend for the northern group (Fig. 3a) and the helium isotope ratio of geothermal fluids at Mt. Pelée overlaps the MORB range [$7.8R_A$ —Fig. 2a and Van Soest *et al.* (1998) and Pedroni *et al.* (1999)], it seems reasonable to include Martinique with the northern islands. However, from Sr, Nd and O isotope results, there is strong evidence for extensive crustal contamination on Martinique (e.g. Davidson, 1987; Davidson & Harmon, 1989). This issue will be addressed further below.

He isotope systematics and mineral chemistry

An important test of the crustal contamination hypothesis is whether there is supportive evidence in addition to the $^3\text{He}/^4\text{He}$ –[He] relationships discussed above. For example, Gasparon *et al.* (1994) showed that $^3\text{He}/^4\text{He}$ ratios correlate with a fractionation index (*mg*-number) in Sunda arc lavas such that phenocrysts with lower *mg*-number, i.e. more evolved lavas, tended to have lower $^3\text{He}/^4\text{He}$ values. This was interpreted to result from degassing during magma evolution with subsequent increased susceptibility of volatile-poor magmas to crustal additions. In this section, we apply the same test to the Lesser Antilles samples.

In Fig. 5, we plot helium isotope ratios against *mg*-number. The data are divided into the two geographical groups distinguished in the previous section. Overall, *mg*-number– $^3\text{He}/^4\text{He}$ data for the Lesser Antilles display the same features as observed in the Sunda arc (Gasparon *et al.*, 1994).

In Fig. 5a, the data for the northern arc are plotted, showing two distinct patterns. The olivine helium isotope data do not show any correlation with *mg*-number, as the ol has MORB-like helium isotope ratios regardless of *mg*-number. In contrast, the px helium isotope data appear to show a positive correlation with *mg*-number. These relationships indicate that (1) assimilation and fractional crystallization (AFC) was an active process in the northern Antilles islands, and (2) assimilation must have been a late-stage feature when ol had already become closed to helium exchange with the host magma.

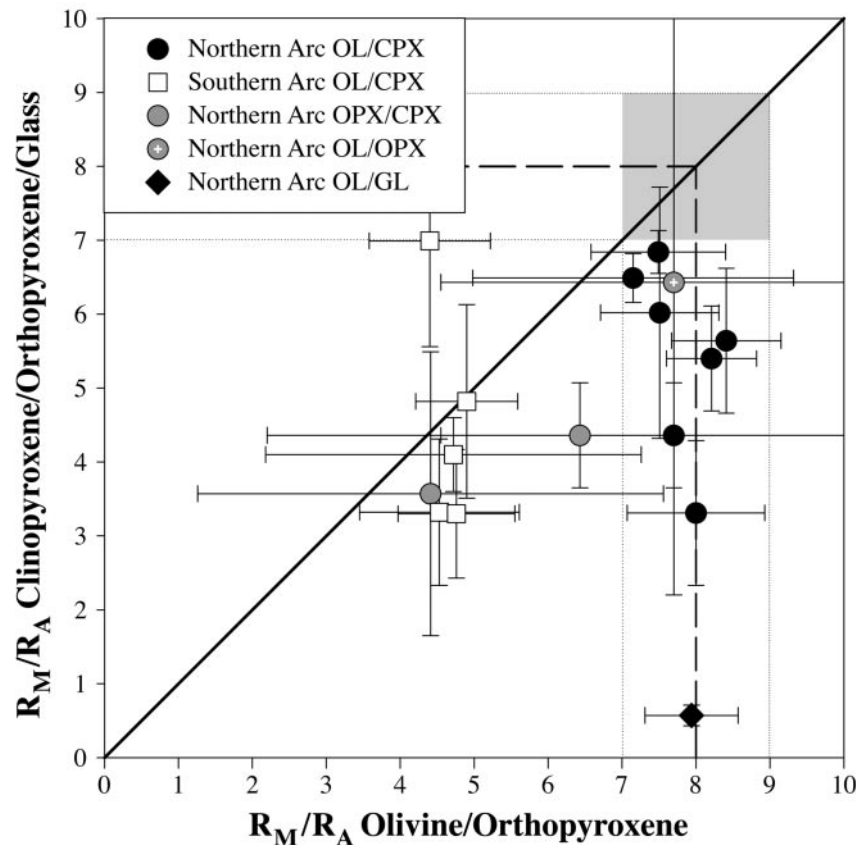


Fig. 4. A plot of helium isotope ratios for cogenetic minerals pairs. Data are divided into groups based on geographical location in the arc and on type of mineral pair. In the case of helium isotope equilibrium samples should fall on the 1:1 line. The figure clearly illustrates the marked difference between the northern and southern arc. When the 2σ error is taken into account, both groups have several mineral pairs that are in equilibrium, but several of the mineral pairs show disequilibrium, which appears greater for the northern arc.

On the basis of proposed liquid lines of descent for Lesser Antilles magmas (e.g. Sigurdsson *et al.*, 1973; Brown *et al.*, 1977; Arculus & Wills, 1980; Devine & Sigurdsson, 1983; Devine, 1995; Heath *et al.*, 1998a), AFC processes take place only in high-level, i.e. shallow-level, crustal magma chambers when ol is already present and no longer plays a significant role as a crystallizing phase.

In Fig. 5b, the data for the southern arc are plotted. It is evident that the helium data show neither trends nor correlations with *mg*-number. Significantly, the two ol samples from Grenada have MORB-like helium isotope ratios and primitive, high *mg*-number—this represents clear evidence that increased amounts of sediment subducted in the southern part of the Lesser Antilles are not necessarily associated with a source of radiogenic helium. However, for the bulk of the southern arc samples, crustal addition has affected both ol and px, reinforcing conclusions drawn in the previous section. The apparent lack of any correlations with *mg*-number in both the olivine and the pyroxene data confirms that late-stage small-degree assimilation following degassing is not the

major factor controlling the helium isotope systematics of these southern arc lavas.

If we consider the single Martinique sample for which a mineral *mg*-number was determined (Fig. 5a and b), it would appear that this sample follows the trend defined by the northern arc samples. The $^3\text{He}/^4\text{He}$ -*mg*-number relationship for Martinique, therefore, supports its classification with the northern islands based upon the $^3\text{He}/^4\text{He}$ -[He] systematics.

The southern Lesser Antilles—a region of pervasive crustal contamination?

In Fig. 5c, we plot the ol $^3\text{He}/^4\text{He}$ against *mg*-number. The plot reinforces the conclusion (previous section) of minimal addition of crustal volatiles in the northern portion of the arc; however, there appear two trends evident for the southern Antilles:

(1) The ‘high *mg*-number’ group is characterized by primitive (high) *mg*-numbers and MORB-like helium isotope ratios. This group is made up of just two samples

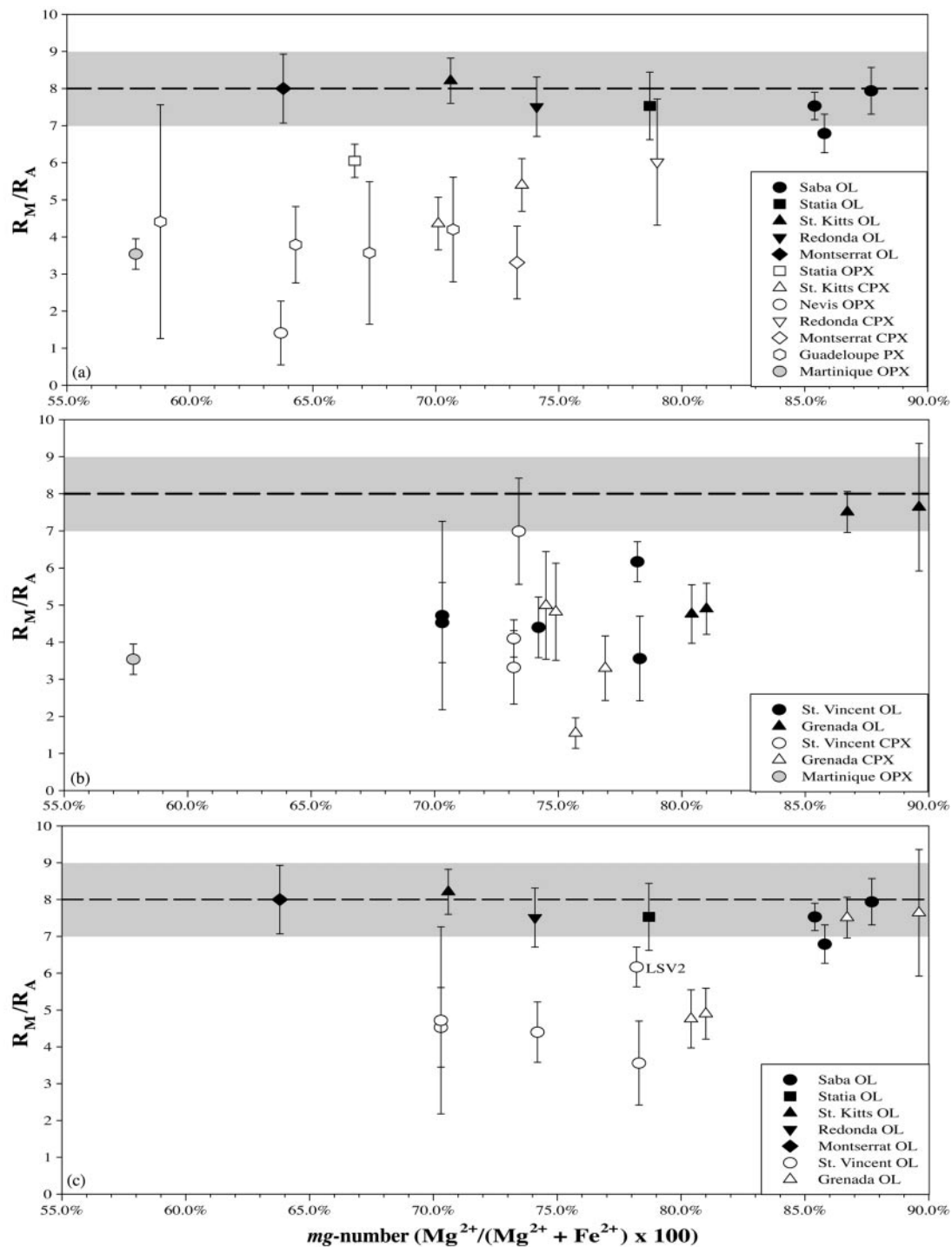


Fig. 5. (a) R_M/R_A vs mg -number for the northern arc. The data are plotted per island and mineral type. Two trends can be observed: (1) ol data for the northern arc do not correlate with mg -number, but instead form a horizontal trend that shows similar $^3\text{He}/^4\text{He}$ independent of mg -number; (2) px data for the northern arc show a broad positive correlation with mg -number, indicating that AFC processes occurring during late-stage degassing have modified their helium isotope systematics. During these events, ol phenocrysts must have been closed for helium exchange with the host magma. (b) R_M/R_A vs mg -number for the southern arc. The data are plotted per island and mineral type. No obvious trends can be observed for the dataset as a whole or for separate mineral type. (c) R_M/R_A vs mg -number for ol phenocrysts. Data are plotted per island and geographical position in the arc. Both sample suites define a more or less horizontal trend of $^3\text{He}/^4\text{He}$ vs mg -number but the northern samples have significantly higher $^3\text{He}/^4\text{He}$. Two samples from Grenada would fit better with the northern arc data.

from Grenada (GRE27x13 and LAG4). Although mineral *mg*-number data are absent, we include the samples from Ile de Caille (WIC17, WIC19, WIC20) with this group based on their $^3\text{He}/^4\text{He}$ (Table 1) and WR geochemistry [reported by Van Soest (2000)].

(2) The 'low *mg*-number' group have lower *mg*-number and $^3\text{He}/^4\text{He} < \text{MORB}$, and are defined by the remainder of the Grenada and St. Vincent samples.

The 'low *mg*-number' group appears to define a trend sub-parallel to the northern arc samples, but displaced to lower helium isotope ratios. It is significant that px from these samples shows little evidence for late-stage assimilation as in the case of px from the northern arc. This feature could be explained by either loss of leverage available for this process—as a result of the presence of earlier crustal addition events, which have already significantly lowered the helium isotope ratios, or closed system fractional crystallization without assimilation. We prefer the former explanation, as widespread evidence for the presence of high-level AFC in the southern islands is reported in the literature (e.g. Hawkesworth & Powell, 1980; Thirlwall & Graham, 1984; Davidson, 1985, 1987; Vidal *et al.*, 1991; Smith *et al.*, 1996; Thirlwall *et al.*, 1996; this work).

The high *mg*-number group has helium isotope ratios that are MORB-like or close to MORB and *mg*-numbers that are high, indicating that crustal additions or AFC have played little part in the history of these magmas. These features could be explained by rapid transit times of the magmas from source to surface. If the magma traverses the arc crust rapidly, there will be little opportunity for large-scale pervasive crustal contamination, open system fractionation (AFC), differentiation or late-stage degassing of these magmas. There are several points that support this scenario:

(1) the samples from Grenada and Ile de Caille (LAG4, WIC17, WIC19, WIC20) are associated with deep-seated fault structures in the arc crust (Westercamp, 1979). These structures can facilitate rapid magma movement by providing a direct conduit to the surface, thus shortening transit times from source to surface.

(2) The high MgO contents of the high *mg*-number group suggest that olivine accumulation may have been an important process for these magmas during their history, e.g. sample LAG4 has almost 18 wt % MgO (Van Soest, 2000). We speculate that to maintain primitive (high *mg*-number) magmas, considerable transit velocities and hence short residence times in crustal magma chambers would be required.

(3) With the exception of GRE27x13 (mantle xenolith), all samples have (slightly) higher helium contents compared with other samples from the southern arc. This might be indicative of reduced opportunity for magma degassing in high-level crustal magma chambers.

For the 'low *mg*-number' group samples, the cumulative helium isotope data (ol, px, and geothermal fluids) suggest the occurrence of at least one major crustal contaminant that has affected magmas early on in the crystallization sequence, before major degassing and 'closure' of olivine to helium exchange with the host magma. This contaminant affects the magmas so extensively that late-stage differentiation and (A)FC processes in high-level magma chambers (as indicated by decreasing *mg*-number of the minerals) have little or no effect on the He isotope systematics.

He–O isotope relationships

Oxygen isotopes can be used as an indicator of crustal contamination, in a manner similar to helium isotopes. Therefore, it is useful to seek relationships between helium and oxygen isotopes in magmatic systems that have undergone differing amounts of crustal contamination. Any kind of relationship will depend on the sensitivity of both isotope systems to crustal contamination events and to potential sources of contamination. In terms of determining source(s) of contamination, oxygen isotopes provide a potentially powerful tool, because many components that can be present in the arc crust have differing $\delta^{18}\text{O}$ values (e.g. Margaritz *et al.*, 1978; James, 1981; Bacon *et al.*, 1989; Feeley & Sharp, 1995; Macpherson *et al.*, 1998).

Our dataset for the Lesser Antilles contains 28 mineral separates that have both helium and oxygen isotope data available (Table 1). In Fig. 6, oxygen isotope data are plotted against helium isotope ratios. There are no general trends evident in the plot but several features also observed in the oxygen dataset stand out and can help to confirm and constrain better the conclusions drawn from the helium and *mg*-number systematics. For example, for the (northern) island of Guadeloupe the sample (lsgu5-px) with the lowest $^3\text{He}/^4\text{He}$ ratio also records a small $\delta^{18}\text{O}$ decrease relative to the anticipated mantle cpx range. The low $^3\text{He}/^4\text{He}$ was interpreted as late-stage assimilation in high-level crustal magma chambers, facilitated by extensive degassing of the magma. The low $\delta^{18}\text{O}$ decrease appears to be consistent with the scenario that as assimilation increases and helium is increasingly modified, oxygen isotopes also start recording the contamination event(s). Modification towards lower $\delta^{18}\text{O}$ values implies that the origin of this contaminant could be either hydrothermally altered magmatic or volcanic wall rocks (e.g. James, 1981; Bacon *et al.*, 1989; Feeley & Sharp, 1995) or hydrothermally altered oceanic crust of the arc basement (e.g. James, 1981; Eiler *et al.*, 1996; Macpherson & Matthey, 1998). To be consistent with our interpretation of the helium isotope systematics of the northern arc px, we suggest that the source of the low

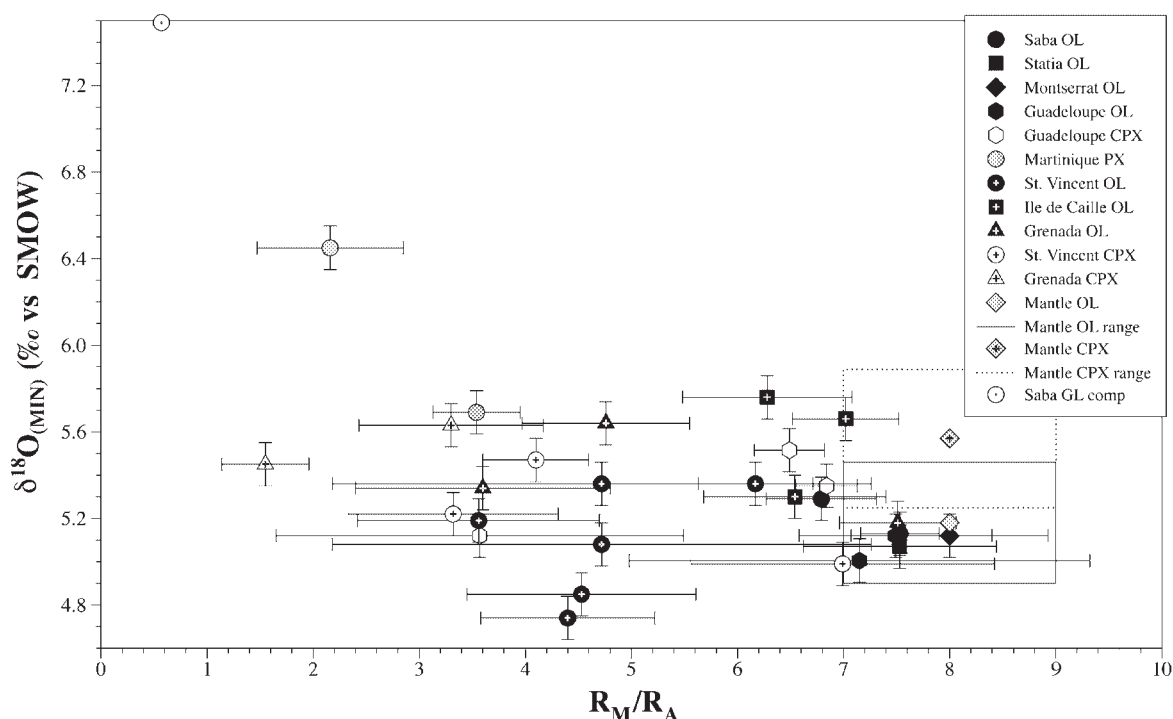


Fig. 6. $\delta^{18}\text{O}$ vs R_M/R_A showing the lack of correlation between helium and oxygen isotope ratios. Most samples show no deviation from mantle oxygen isotope values (shown by the dotted-line box for px and the continuous-line box for ol), whereas helium isotopes record crustal contamination. The northern glass represents a data composite from the two glasses from the island of Saba; helium isotope data were obtained for one sample whereas oxygen isotope data were obtained for the other. For display purposes we assume that He and O are interchangeable for these samples, as other geochemical data are similar [see Table 1 and Van Soest (2000)].

$\delta^{18}\text{O}$ is hydrothermally altered magmatic or volcanic wall rocks situated at high levels in the arc crust.

In the southern portion of the arc, the oxygen isotope data do not show any correlation with the two groups distinguished based on helium isotope–*mg*-number systematics. The bulk of the data falls within the oxygen isotope range for mantle peridotite, in contrast to the helium isotope data, which show clear evidence for crustal contamination. This decoupling of He and O isotope systematics is probably due to a difference in sensitivity to crustal contaminants. The ‘low *mg*-number’ group (in Fig. 6 mainly defined by $^3\text{He}/^4\text{He}$ ratios $<5R_A$) includes some samples (e.g. from St. Vincent) that show oxygen isotope shifts towards lower ratios. On the basis of the previous discussion [suggesting a pervasive (deep) crustal source of contamination], it could be argued that the source for the contaminant in this case is not restricted to high-level crustal magma chambers and therefore could be altered oceanic crust of the arc basement. However, the fact that helium and oxygen isotope systematics are not necessarily related also allows for an alternative interpretation. The St. Vincent samples showing modifications towards lower oxygen isotope ratios are related to recent eruptions of the Soufrière volcano. Until three decades ago, this volcano was characterized

by the presence of a large crater lake (e.g. Robson & Tomblin, 1966; Devine & Sigurdsson, 1983), which indicates that large amounts of meteoric water are potentially available to circulate through the uppermost arc crust and participate in hydrothermal alteration.

In addition, there are three olivine samples in the southern arc that have high $\delta^{18}\text{O}$ values compared with olivine in equilibrium with mantle peridotite (Mattey *et al.*, 1994). This indicates that there is a contaminant present in the arc crust with high $\delta^{18}\text{O}$ values. The origin of this contaminant could be several types of sediment incorporated in the crust or the arc crust itself (e.g. Margaritz *et al.*, 1978; James, 1981). The observation that two of these olivine samples fall within the ‘high *mg*-number’ group implies that oxygen contamination can occur independently of helium contamination or that the contaminant changes in composition.

The sample that shows the most convincing evidence for crustal contamination in oxygen isotopes is a pyroxene sample from Martinique—an island where crustal contamination has been well documented by previous $\delta^{18}\text{O}$ studies of WR and plagioclase separates from the older centers (Davidson, 1985; Davidson & Harmon, 1989). The high $\delta^{18}\text{O}$ pyroxene (lsm2) in this study comes from one of these centers. The other sample from Martinique,

which shows no evidence for crustal contamination in its oxygen isotopes (lsm4b), was erupted from Mt. Pelée, a center where crustal contamination is much less apparent and of minor importance (e.g. Davidson, 1986, 1987).

He–Sr isotope relationships

The strontium isotope record of arc-related rocks potentially represents a complex history of enrichment by subducted components in the source region as well as later modification by crustal magma chamber processes. Indeed, there is evidence for both processes in the Sr isotope record of Lesser Antilles lavas (e.g. Hawkesworth & Powell, 1980; Davidson, 1983; Thirlwall & Graham, 1984; Davidson, 1985, 1987; White & Dupre, 1986; Vidal *et al.*, 1991; Thirlwall *et al.*, 1996; Turner *et al.*, 1996; Heath *et al.*, 1998a). Again, therefore, it is of interest to consider isotope–isotope correlations, in this case between He and Sr, to gauge relative sensitivities to these processes. It should be noted, however, that lavas in which the most convincing evidence for crustal contamination is found are intrinsically unsuited for He isotope studies as a result of their more evolved compositions.

In an attempt to quantify the effects of crustal contamination on both He and Sr isotopes, we consider our data in terms of end-member compositions (Lesser Antilles arc crust, subducting material, and the mantle wedge) and possible mixing relationships between them (after Langmuir *et al.*, 1978). Table 2 details the end-member compositions used in the modeling. We make the assumption that WR Sr-isotope ratios are representative of values in the phenocrysts: initial work comparing WR with ol and px Sr isotope ratios (Van Soest, 2000) indicates this is the case for the majority of samples considered here. We consider two scenarios: (1) mixing between subducted components and mantle wedge; (2) admixture of variably degassed arc magma with arc crust.

In Fig. 7, we plot the total He–Sr isotope dataset for the Lesser Antilles superimposed upon various mixing trajectories between the MORB source and the end-members described above [scenario (1)]. We point out that for these mixing scenarios our subducted end-members are assumed to retain their helium during subduction. We note that if helium is not subducted, which is the most likely scenario for ‘normal’ subduction not involving continental crust (Staudacher & Allègre, 1988; Hilton *et al.*, 1992; Hiyagon, 1994a, 1994b), then subduction addition would be reflected only in the Sr composition of samples: this is illustrated in Fig. 7 by the dashed lines extending horizontally from the MORB source (MS) box towards higher Sr isotope ratios.

With this in mind the following points are apparent from Fig. 7:

(1) most of the data points plot far away from the mixing curves calculated for the subduction addition scenario. This implies that subduction of helium in the subducting slab and sediments cannot explain the bulk of the data, as all sedimentary and the altered MORB end-members have been defined assuming complete retention of their helium through the subduction process.

(2) The Sr isotopic composition of the sediment end-members is of little consequence, as the GL (GLOSS), SN (northern arc sediment), and SSA (southern arc sediment—alternate) mixing curves follow essentially the same trend for the Sr isotope range (up to 0.7055) covered by the data of this study.

(3) The MS (MORB source) \times AM (altered MORB) mixing curve represents mixing of MORB source with a subducted component without involvement of sediment melt or slab-derived fluid. Because most data points plot well away from this mixing curve, it is clear that mixing with a component derived from altered MORB alone cannot explain the data for the Lesser Antilles. Most data points plot away from the MS \times AM trajectory and at least require the addition of a sediment component either as a distinct melt [as described by Elliott *et al.* (1997)] or as a contributor to the slab-derived fluid to explain their Sr isotope composition.

(4) The northern and southern groups have distinct Sr isotope characteristics. The northern group defines a range between 0.7031 and 0.7038 and the southern group defines a range from 0.7039 to 0.7054. On the basis of the Sr isotope results alone, this clear difference between the north and the south can be explained simply by adding a greater proportion of a subducted sediment component in the south compared with the north. The helium isotope results require that some amount of crustal addition is involved as described above.

(5) Northern group ol data can be explained by subduction addition alone; only the Sr and not the helium isotopes have been affected. Northern group px data have been affected by minor late-stage crustal contamination (see the discussion for helium systematics above); whether or not this has also affected the Sr isotopes cannot be determined at this time, but it would appear to be not the case (see below).

(6) For the southern group, all but three of the data points fall below the dashed box defining subduction addition (no helium subduction). This indicates that most of the samples have been affected by crustal contamination. The range of $^{87}\text{Sr}/^{86}\text{Sr}$ for the three samples that could be explained by only subduction addition spans almost the whole range observed for the southern group in this study. This indicates that for Sr isotope ratios all the data points could essentially be obtained by subduction addition only.

In scenario (2) given above, we consider the case of admixture of variably degassed arc magma with arc crust

Table 2: Sr–He mixing end members and *K* values

End member: ^a	[⁴ He] (ncm ³ STP/g)	R _M /R _A	[Sr] (ppm)	⁸⁷ Sr/ ⁸⁶ Sr	ID ^b	Subduction mixing curves ^c	<i>K</i> ^d	Crustal mixing curves ^e	<i>K</i> ^d
MORB source	5000	8.00	9.0	0.702730	MS	MS × GL	60.56	MAN × CN	0.13
GLOSS	3000	0.05	327.0	0.717300	GL	MS × SN	47.44	MAN– × CN	0.17
Sed. N	1300	0.05	111.0	0.717880	SN	MS × SS	14.42	MAN+ × CN	0.10
Sed. S	5200	0.05	135.0	0.711000	SS	MS × SSA	42.74		
Alt. MORB	390	0.05	120.0	0.703100	AM	MS × AM	170.94	MAS1 × CS1	0.001
Sed. S Alt.	5200	0.05	400.0	0.720000	SSA			MAS1 × CS2	0.013
MORB ARC N	10	8.00	200.0	0.703494	MAN			MAS1 × CS3	0.200
MORB ARC N–	10	8.00	150.0	0.703200	MAN–				
MORB ARC N+	10	8.00	250.0	0.703800	MAN+			MAS2 × CS1	0.003
MORB ARC S1	10	8.00	300.0	0.703900	MAS1			MAS2 × CS2	0.027
MORB ARC S2	20	8.00	300.0	0.705500	MAS2			MAS2 × CS3	0.400
MORB ARC S3	20	8.00	150.0	0.703900	MAS3				
CRUS N	100	0.05	250.0	0.704500	CN			MAS3 × CS1	0.005
CRUS S1	10000	0.05	400.0	0.711000	CS1			MAS3 × CS2	0.053
CRUS S2	1000	0.05	400.0	0.720000	CS2			MAS3 × CS3	0.800
CRUS S3	100	0.05	600.0	0.711000	CS3				

^aThe end members are defined as follows. MORB source: undergoing 10% of partial melting; [Sr] is modified after Sun & McDonough (1989) and [He] is modified after Jambon *et al.* (1985) and Sarda & Graham (1990); GLOSS: Sr from Plank & Langmuir (1998), [⁴He] is an estimate based on results for the Lesser Antilles sediments (Van Soest, 2000); Sed. N (subducted sediments for the northern arc): Sr from Plank & Langmuir (1998), He from Van Soest (2000); Sed. S (subducted sediments for the southern arc): Sr from Plank & Langmuir (1998), He from Van Soest (2000); Alt. MORB (subducted altered oceanic crust): Sr from White *et al.* (1985), He from Van Soest (2000); Sed. S Alt.: an alternative composition for subducted sediment in the southern arc, based on more extreme compositions found for Southern Lesser Antilles sediments (White *et al.*, 1985; Ben Othman *et al.*, 1989); MORB ARC N, N–, N+: potential composition of arc magmas in the northern Lesser Antilles as they enter the arc crust after a first stage of deep degassing, based on results from this study; MORB ARC S1, -2, -3: potential composition of arc magmas in the southern Lesser Antilles as they enter the arc crust after a first stage of deep degassing, based on results from this study; CRUS N: best estimate for the composition of a northern Lesser Antilles arc crustal component; CRUS S1, -2, -3: several potential compositions of southern Lesser Antilles arc crustal components, based on the presence of sediments in the crust that are similar to those subducted beneath the arc.

^bEnd-member identifier.

^cMixing curves calculated for the mixing of subducted components with the MORB source.

^d*K* values for the various mixing curves. The *K* value is calculated based on the He and Sr contents of the components that are mixing: $K = ([^4\text{He}]_{\text{Mxx}} \times [\text{Sr}]_{\text{cont}}) / ([\text{Sr}]_{\text{Mxx}} \times [^4\text{He}]_{\text{cont}})$, where Mxx represents any of the mixing components starting with an M, and cont represents the other mixing components, which are in effect the contaminants added.

^eMixing curves calculated for the mixing of the arc magma as it enters the arc crust with potential crustal components.

as the He isotope data indicate that there must be a component of crustal addition. To evaluate this possibility, mixing calculations have been carried out for a set of arc magmas, after transfer from their source, which have undergone subduction contamination and an initial stage of degassing, with a set of potential crustal end-members thought to be appropriate for the Lesser Antilles. The end-member compositions and related *K* values are given in Table 2.

In Fig. 8, we plot the He–Sr data superimposed upon crustal addition mixing curves. There is one set of mixing curves for the northern arc based on three potential

magmas mixing with one crustal component. For the southern arc there are three sets of mixing curves labeled A, B, and C. The curves are based on three magmas (MAS1–3) mixing with three crustal contaminants (CS1–3).

The following points are apparent:

(1) the mixing curves calculated for the crustal contamination in the northern part of the arc are consistent with the scenario of subduction addition with some late-stage crustal addition. The late-stage contamination has apparently not affected the Sr isotopes. The Sr isotope systematics of the northern group are determined by the

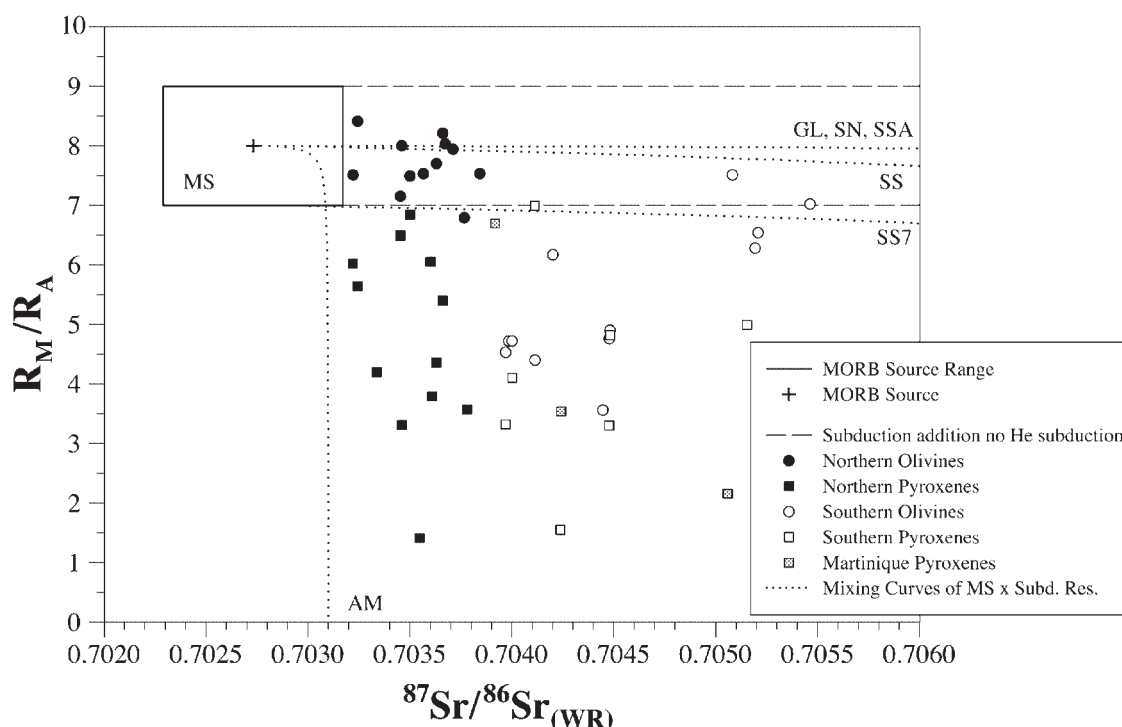


Fig. 7. $^3\text{He}/^4\text{He}_{(\text{MIN})}$ (R_M/R_A) vs $^{87}\text{Sr}/^{86}\text{Sr}_{(\text{WR})}$ with mantle–subducted component mixing curves superimposed (subduction mixing curves). The mixing curves represent an end-member scenario with the assumption of complete He retention in the subducted components. The alternative scenario is presented by the dashed-line box extending from the MS box towards higher $^{87}\text{Sr}/^{86}\text{Sr}$, labeled ‘Subduction addition no He subduction’. This represents the case where no He survives the subduction process and subduction addition is reflected only in Sr isotopes. All data points fall to the right of the MS \times AM mixing curve, indicating the involvement of subducted sediment in all cases. The northern group of data can be explained by subduction addition only but most of the other data points fall below the mixing curves defining subduction addition, indicating some extent of crustal contamination.

subducted component after which late-stage contamination affected the helium isotope systematics of the px. This implies that K values must be very low, probably less than the minimum of 0.1 calculated here, and gives the appearance that He and Sr behave independently. However, as discussed by Hilton *et al.* (1992), this is mainly due to the relatively large He/Sr ratios in the contaminants, which give helium a greater sensitivity to crustal contamination compared with strontium.

(2) For MAS \times CS mixing curves with low K values (curves labeled A) the Sr isotope composition of the CS end-members is of little consequence for defining trends: all three curves have similar curvature and indicate that crustal addition initially affects only helium isotope systematics without significant changes to Sr isotopes. Only in the case of mixing curves with K values close to unity, i.e. fairly straight mixing lines (curves labeled B), does the isotopic composition for the CS end-members have an important influence on resulting values of He and Sr isotopes.

(3) As noted above, the He isotope data for all but three samples of the southern group suggest the presence

of crustal contamination. This contamination has affected ol as well as px and implies a process that is distinct from that occurring in the northern arc.

For the southern arc, two ‘end-member’ scenarios are compatible with the mixing curves presented in Fig. 8:

(1) subduction addition alone accounts for the observed Sr isotope ratios. The subsequent crustal addition events, as recorded in the He isotope results, can then be attained by adding He following mixing curves with very low K values ($K < 0.01$; see the curves marked A in Fig. 8), so that Sr is unaffected. This scenario implies again that He appears to behave independently of Sr with respect to tracing crustal addition.

(2) If most of the southern arc magmas do not have Sr isotope ratios much higher than the composition of MAS1,-3 then subsequent ^{87}Sr enrichment is achieved through crustal addition along mixing curves marked B with K values close to unity. If the potentially large range in Sr isotope ratios for the crustal end-member in the south is taken into account (0.711–0.720; see Table 2), it is possible to explain all the data in the southern arc with these types of mixing relationships.

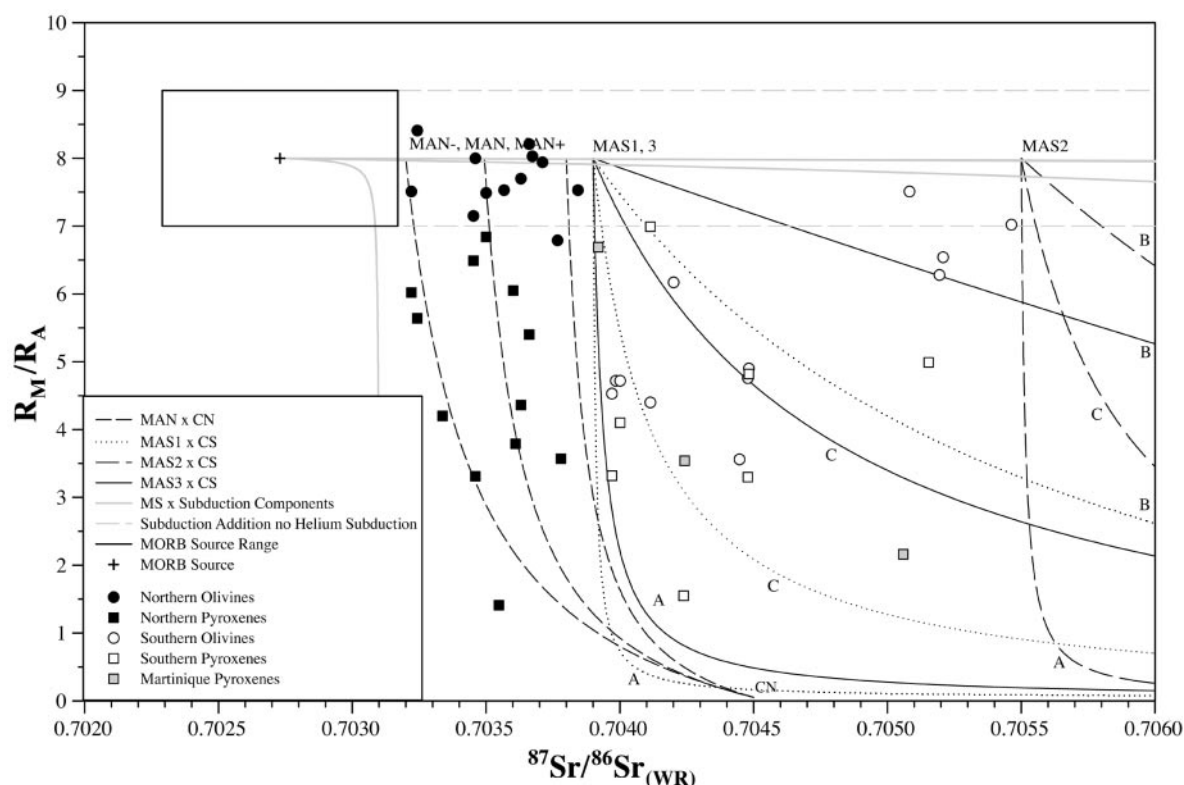


Fig. 8. $^3\text{He}/^4\text{He}_{(\text{MIN})}$ (R_M/R_A) vs $^{87}\text{Sr}/^{86}\text{Sr}_{(\text{WR})}$ showing arc magma–crustal component mixing curves (crustal mixing curves). For the northern arc, there is one set of three mixing curves between three potential arc magma end-members and one crustal end-member, which represents the northern arc crust (see Table 2). For the southern arc, there are three sets of three mixing curves: three sets of potential southern arc magma end-members (forming two starting points: MAS1, 3 and MAS2) mix with three sets of potential southern arc crustal end-members (see Table 2). The mixing curves from Fig. 7 are shown in the background in gray. Mixing curves marked A have K values <0.01 and represent dominance of He in the crustal component. Mixing curves marked B have K values closer to unity (>0.1) and represent cases where Sr isotopes play a major part in the crustal addition scenario. Mixing curves marked C have K values $0.01 < K < 0.1$ and represent a combination of previous possibilities.

Arguments in favour of scenario (1) are the increased amounts of sediment subducted in the south of the arc (von Huene & Scholl, 1991; Plank & Langmuir, 1998), which could lead to high Sr isotope ratios by subduction addition alone. Curves marked C in Fig. 8 represent possibilities where the strontium isotope signature of some samples is generated by crustal contamination. This implies that mixing curves with characteristics similar to those of curves A ($K < 0.01$) and C represent plausible mixing scenarios; in which case, the main shift in Sr isotopes is caused by subduction addition.

It is not yet possible to evaluate the validity of scenario (2), because of the difficulty of constraining the starting Sr isotope composition of the magmas as they enter the arc crust. However, if scenario (2) represents a realistic possibility, some form of Sr isotope disequilibrium between the different minerals might be expected. Preliminary experiments carried out on some of the samples (Van Soest, 2000) suggest that this is not the case, and therefore we would argue against this scenario. We

conclude, therefore, that when there is clear evidence from Sr isotopes and/or other geochemical parameters for crustal additions, helium will be expected to give corroborating evidence. However, when there is clear evidence for crustal additions in the He isotope system, Sr isotopes (and/or other geochemical parameters) will not necessarily match, because of the greatly increased sensitivity of helium to crustal contamination.

Sr–O isotope relationships

Sr–O relationships have been used frequently to identify the presence of crustal contamination in magmas (e.g. Margaritz *et al.*, 1978; James, 1981; Harmon *et al.*, 1984; Davidson, 1985; Bacon *et al.*, 1989; Davidson & Harmon, 1989; Harris, 1989; Smith *et al.*, 1996; Thirlwall *et al.*, 1996; Macpherson *et al.*, 1998). Indeed, for the Lesser Antilles, correlations between WR $\delta^{18}\text{O}$ values and Sr, Nd and Pb isotopes are consistent with extensive crustal

contamination of the lavas of Martinique (e.g. Davidson & Harmon, 1989). Subduction contamination produces only minor changes in oxygen isotope ratios (e.g. Macpherson & Mattey, 1998; Macpherson *et al.*, 1998; Vroon *et al.*, 1999; Eiler *et al.*, 2000) so large shifts in oxygen isotopes from mantle values are most likely to represent crustal contamination. On the other hand, increases in Sr isotopes without modification of oxygen isotopes probably point to subduction contamination.

In Fig. 9, we plot $^{87}\text{Sr}/^{86}\text{Sr}$ vs $\delta^{18}\text{O}$ for both the northern and southern portions of the arc (Table 1) together with literature data (available only for the southern arc). Superimposed on the plot are several mixing curves. The northern arc shows a number of features noted previously; for example, most data can be explained by subduction contamination only (curve labeled SC). However, the deviation from mantle $\delta^{18}\text{O}$ values for three samples is best explained by contamination with arc crust. For shifts towards low $\delta^{18}\text{O}$, the arc crust would be hydrothermally altered (e.g. James, 1981; Bacon *et al.*, 1989; Feeley & Sharp, 1995); if deviations are towards high $\delta^{18}\text{O}$ ($>7\text{‰}$), as illustrated by the glass sample (curve NA: $\delta^{18}\text{O} = 10$; $^{87}\text{Sr}/^{86}\text{Sr} = 0.704500$), then the contaminant is probably arc crust that has not been hydrothermally altered (Margaritz *et al.*, 1978; Longstaffe *et al.*, 1983; Harmon *et al.*, 1984; Davidson & Harmon, 1989). In either case, significant shifts in $^{87}\text{Sr}/^{86}\text{Sr}$ are not expected.

For the southern arc, there are two crustal contamination trends in oxygen isotopes: a trend towards lower $\delta^{18}\text{O}$ values (identified by two arrows in Fig. 9) identifies hydrothermally altered crust, as for the northern arc. For the trend towards higher $\delta^{18}\text{O}$ values, we suggest that the $^{87}\text{Sr}/^{86}\text{Sr}$ data may help constrain mixing trends better. The starting composition for potential mixing curves falls somewhere along the SC mixing curve (Fig. 9), as subduction contamination affects mostly Sr and not O isotopes. The data are then constrained by mixing curves that trend towards the composition of local sediments [$\delta^{18}\text{O} \sim +20\text{‰}$ (Davidson, 1987) and $^{87}\text{Sr}/^{86}\text{Sr}$ 0.711–0.720 (White *et al.*, 1985; Plank & Langmuir, 1998)], which have been identified previously as being the possible source of crustal contamination in the southern arc (e.g. Davidson & Harmon, 1989; Thirlwall *et al.*, 1996). In contrast to He/O and He/Sr systematics, the Sr/O of magma and contaminant will be fairly similar thereby giving K values close to unity, where $K = ([\text{Sr}]_{\text{m}}/[\text{O}]_{\text{m}})/([\text{Sr}]_{\text{c}}/[\text{O}]_{\text{c}})$, which simplifies to $K = [\text{Sr}]_{\text{m}}/[\text{Sr}]_{\text{c}}$, as in most cases $[\text{O}]_{\text{m}} \sim [\text{O}]_{\text{c}}$ (where m is magma and c is contaminant). Such mixing scenarios are illustrated by curves SA1–SA4 ($\delta^{18}\text{O} \sim +20\text{‰}$ for all; $^{87}\text{Sr}/^{86}\text{Sr}$ 0.711 for SA1, SA3, CC1, and CC2; $^{87}\text{Sr}/^{86}\text{Sr}$ 0.720 for SA2 and SA4). The data for the southern arc, including literature data, generally conform to this set of mixing trajectories. However, mixing curves following the trends

to end-members CC1 and CC2, indicating Sr modification by subduction contamination followed by O modification by crustal contamination, are unrealistic based on the anticipated end-member Sr/O.

Noble gases in Martinique: major crustal addition or late-stage degassing?

The island of Martinique plays a pivotal role in the discussion about crustal additions in the Lesser Antilles. Geochemical studies (e.g. Davidson, 1986; Davidson & Harmon, 1989; Smith *et al.*, 1997) have shown evidence for the presence of significant crustal additions to magmas with the contaminant probably of sedimentary origin. However, helium isotopes on Martinique present a contrasting image, with $^3\text{He}/^4\text{He}$ ratios of $7.8R_{\text{A}}$ for a geothermal fluid from Mt. Pelée (Van Soest *et al.*, 1998; Fig. 2a) and similar MORB-like values in geothermal fluids for the Diamant volcanic center (Pedroni *et al.*, 1999). Helium isotope results of phenocrysts (this work) resemble data for the northern Lesser Antilles (Figs 3 and 5), where there is little evidence for the presence of extensive crustal additions. This appears to be confirmed by the fact that the oxygen isotope composition of the orthopyroxene separate (lsm4b; Table 1) does not deviate from mantle values. The helium isotope ratio of $3.43R_{\text{A}}$ for this orthopyroxene can probably be related to late-stage assimilation following degassing, as is thought to occur in the northern arc.

The other pyroxene sample (lsm2) for Martinique for which helium, oxygen and strontium isotope data are available is from an older center, Pitons du Carbet (>0.6 Ma; Nagle *et al.*, 1976; Briden *et al.*, 1979). Even though its helium isotope ratio ($2.16R_{\text{A}}$) must be considered a minimum value, its oxygen and strontium isotope characteristics are consistent with the presence of a major crustal contamination component (Figs 6 and 9). The fact that the oxygen and strontium isotope characteristics of this sample are among the most extreme values determined during this study confirms observations (e.g. Davidson, 1986; Davidson & Harmon, 1989; Smith *et al.*, 1997) that crustal contamination plays an important role in older centers of Martinique.

Pedroni *et al.* (1999) explained the trend between He and Sr isotopes for Martinique (Fig. 8) by subduction contamination by helium. They compared helium isotope ratios from present-day geothermal fluids with published WR strontium isotope ratios for various volcanic centers of Martinique. We question the validity of this approach given that the $^{87}\text{Sr}/^{86}\text{Sr}$ database was produced using lavas from different volcanic centers over various time periods (e.g. several centers have an age of >10 Ma). For example, it seems improbable to us that the volcanic rocks of the Lamentin center, which show a range in

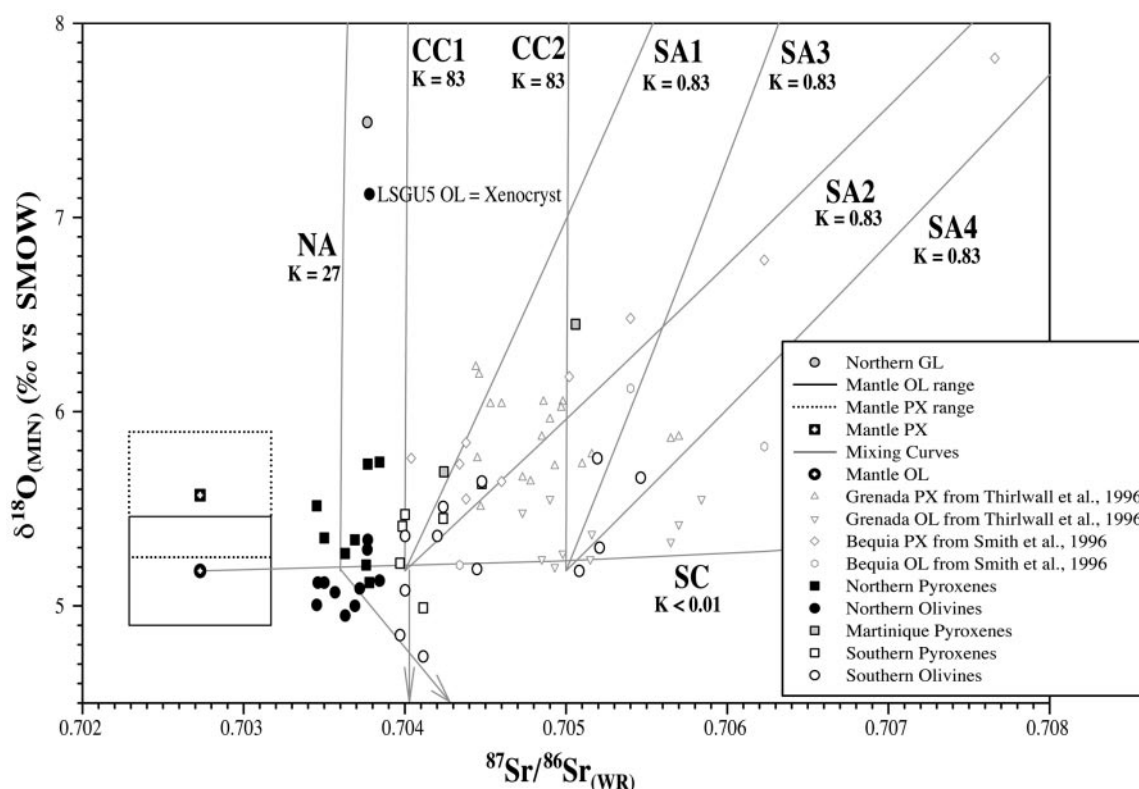


Fig. 9. $\delta^{18}\text{O}_{(\text{MIN})}$ vs $^{87}\text{Sr}/^{86}\text{Sr}_{(\text{WR})}$ showing the dataset divided per group and mineral phase together with literature data for the islands of Bequia (Grenadines) and Grenada (Smith *et al.*, 1996; Thirlwall *et al.*, 1996). Noteworthy features are the clear distinction between the northern and southern segments of the arc, and the two distinct trends for the southern group towards high and low oxygen isotope end-members.

strontium isotope ratios from 0.706 to 0.710 (Davidson, 1986), could be related to a single geothermal system characterized by a present-day helium isotope ratio of around $2R_A$. Contrary to the suggestion that helium is derived from the subducted slab (Pedroni *et al.*, 1999), we interpret the data for the southern arc as showing the effects of crustal contamination as recorded in both the helium and oxygen isotope systematics. Other samples that show features related to subduction contamination are characterized by MORB-like helium isotope ratios with strontium isotope values that can cover a considerable range (Fig. 7 or 8).

To explain the occurrence of both crustal contamination and subduction contamination in lavas from Martinique, its geographical position within the Lesser Antilles must be considered. In and around Martinique, two important changes take place in the Lesser Antilles arc:

(1) north of the island of Martinique, the Lesser Antilles arc is bifurcated: an older inactive set of islands lies to the east and currently active younger islands are located to the west. This shift in activity from east to west

has also shifted volcanism on Martinique itself, thereby moving the current line of volcanism off the central axis of the arc and to the west.

(2) Around the island of Martinique, the fore-arc becomes thinner and smaller in lateral extent (Fig. 1). Therefore, the distribution of the crustal contaminant (sediments) may be more limited compared with the southern Lesser Antilles.

The shift of magmatic activity away from the axis of the arc and the potentially diminished distribution of the sedimentary contaminant may provide an explanation for the absence of discernible crustal additions at the Mt. Pelée center as opposed to the older centers. This suggestion is supported by results from radiogenic isotope studies (e.g. Davidson, 1986; Davidson & Harmon, 1989), which show that evidence for crustal addition is mainly apparent in lavas from the older centers, whereas the younger centers have radiogenic isotope systematics that are similar or close to those of the northern Lesser Antilles (e.g. Hawkesworth & Powell, 1980; Davidson, 1983, 1987; White & Dupre, 1986). Martinique is then the locus of the transition in the Lesser Antilles arc from the

northern islands, where crustal additions play a relatively minor role in petrogenesis, to the southern arc, where crustal contamination is much more extensive.

CONCLUSIONS

In conclusion, the following points are emphasized:

(1) helium isotope and abundance systematics of ol and px phenocrysts from Lesser Antilles arc lavas confirm the observations from helium isotope systematics in geothermal fluids (Van Soest *et al.*, 1998) that there is a regional control on the importance of the role of crustal contamination in petrogenesis. The islands towards the north of the arc (from Martinique northwards) show evidence for only minor late-stage contamination. The islands from the southern arc (Grenada–Martinique) show evidence for a more pervasive, major crustal contamination.

(2) As pyroxene is the only phenocryst phase to record He isotope evidence of crustal contamination in the north, we suggest that the contaminant is probably older arc crust. The crust contributes relatively minor amounts of radiogenic helium to the magma at a late stage—following extensive magmatic degassing when olivine phenocrysts have ceased to exchange helium with the host magma. The fact that both olivine phenocrysts and geothermal fluids record crustal contamination in the south of the arc leads us to suggest that a contaminant other than older arc crust is required to supply the radiogenic helium. We opt for terrigenous sediment, originating either from the older Aves fore-arc or possibly as overthrust sediments from the current fore-arc, as the contaminant.

(3) The only coupled behaviour between tracers of crustal contamination in the Lesser Antilles is between $^3\text{He}/^4\text{He}$ and mg -number. $\delta^{18}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ show similar trends in their geographical distribution, potentially indicative of major crustal contamination in the south, but no clear correlation with He isotope systematics. This is probably because $^3\text{He}/^4\text{He}$ is more sensitive to crustal contamination than $\delta^{18}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$. Within He–Sr isotope space, there are some indications that the complete range in $^{87}\text{Sr}/^{86}\text{Sr}$ for the southern islands could have been caused by subduction contamination alone. However, this scenario cannot be resolved from crustal contamination processes, as indicated by helium isotope systematics, that also cause increases in $^{87}\text{Sr}/^{86}\text{Sr}$.

(4) Sr–O isotope mixing relationships show anticipated trends: subduction contamination is indicated by an increase in $^{87}\text{Sr}/^{86}\text{Sr}$ without a significant shift in $\delta^{18}\text{O}$. In contrast, crustal contamination is indicated by shifts in both $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{18}\text{O}$, which is best modeled by mixing curves with K values close to unity. For the southern group of islands, the mixing trends suggest that

sediments are the most likely candidate to serve as the crustal contaminant.

(5) The island of Martinique appears to be transitional between the two geographic groupings. The recent volcanic centers (Mt. Pélee and Diamant) show little evidence for crustal contamination. Crustal contamination recorded in the literature (Davidson, 1986; Davidson & Harmon, 1989) covers mainly the older centers. This can be explained by a shift in the location of the volcanic arc towards the west of the island.

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